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Analytical Results of Physics Specimens and Dosimeters in Fuel Pins 1, 2, and 4, Irradiated in the Dounreay Prototype Fast Reactor

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FOR THE UNITED STATES
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Date Published: December 1994

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MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-84OR21400

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ACKNOWLEDGMENTS

We thank P. B. Hemmig (USDOE), K. M. Swanson (UKAEA), C. Brown (UKAEA), and T. Mukaiyama (JAERI) for their interest in this work, coöperation, and tangible support. This report was read critically by B. D. Murphy (ORNL) and T. D. Newton (UKAEA).

We express our appreciation to the many people of the Analytical Chemistry Division who contributed to the dissolution and preparation of our samples. A special thanks to M. W. Haskew for his contributions. Thanks also to J. A. Carter, D. A. Costanzo, J. M. Peele, C. A. Pritchard, and others who guided, advised, or analyzed these difficult samples. We also thank Doris C. Smith and Ann M. McCoy for their untiring efforts in preparing the reports and being so tolerant of us in making changes, corrections, and rewrites. Our thanks also to D. H. Smith and R. W. Sharpe for their help in technical proofreading.

ABSTRACT

The United States and the United Kingdom have been engaged in a joint research program in which samples of higher actinides were irradiated in the 600-MW Dounreay prototype fast reactor in Scotland. Three separate fuel pins (FPs) were prepared and irradiated. The actinides in FP-1 and FP-2 were irradiated for 63 full power days (FPD). The irradiation of FP-4 was carried out over a longer period (492 FPD) and should provide the best estimate for cross-section and fission-yield measurements made to date. This report presents the analytical results using mass spectrometry and radiometry for the actinides and the primary activation products for the three FPs. This report also details the fission-product yield measurements for samples of FP-4 by gamma-ray assay techniques with selected results from similar measurements previously obtained for FP-1 and FP-2 samples.

1. INTRODUCTION

The objective of the joint U.S.–U.K. program has been to investigate the neutronic and irradiation behavior of higher actinides in fast reactors. This summary report details the analytical methodologies used to quantify the actinides after irradiation and presents results obtained for the reaction products for the three physics-specimen fuel pins (FPs). Each FP contained dosimeter materials to aid in flux calculations; these results are also presented.

The irradiations were carried out over a period of six years. The total exposure was 295,000 MWd consisting of 38,000 MWd (July 82-Sept. 83), 57,000 MWd (Dec. 85-May 86), 81,000 MWd (Aug. 86-Feb. 87), and 119,000 MWd (Sept. 87-July 88). The total fluence was ~2×10^{23}n/cm^2. A detailed reactor power history as it relates to the exposure of FP-4 will be described in a future report.

Isotopic and material concentration measurements of the target actinides and those created by the irradiations were performed for FP-1 in late 1984, for FP-2 from late 1986 to early 1987, and for FP-4 from mid 1990 to early 1992. (Additional cross checks and remeasurements of FP-4 samples continued up to early 1994.) The procedure involved, first, the cutting away of the outer pin cladding to free individual capsules, and next, the identification and complete dissolution of each capsule separately. These measurements were performed using mass spectrometry and/or radiometry (see Sect. 3 and Appendix A). Appendixes B and C present the summary data sheets for FP-1, FP-2, and FP-4.

Earlier Oak Ridge National Laboratory (ORNL) reports published in this effort detail the preparation of samples for irradiation,[1] characterization of the actinide and dosimeter samples,[2] the preanalysis calculations for FP-1,[3] fission-product yield data,[4] and analyses of samples in FP-1 and FP-2.[5]

2. PHYSICS SPECIMEN MATERIALS AND DOSIMETERS IRRADIATED IN THE U.S-U.K. HIGHER ACTINIDE EXPERIMENT

Twenty-one actinide materials were selected for use as physics specimens for the U.S.–U.K. actinide experiment. Table 1 shows the specimens and their enrichment before irradiation along with their positioning in the three FPs. The vanadium-encapsulated materials were loaded into each FP in five sections (see Fig. 1).

Table 1. Physics specimen materials selected for irradiation

Isotope	Batch No.	Position in FP	Isotope enrichment (wt %)
^{248}Cm	1117	4	92.15
^{246}Cm	C59SHIP	5, 6	66.53
^{237}Np	24HP	7	99.99
^{244}Cm	C57CM46	8, 9	92.35
^{243}Cm	1011	10	57.67
^{238}U	Q1	11	99.955
^{243}Am	H1P1018	12, 13	99.987
^{241}Am	79AMB4	14, 15	99.995
^{242}Pu	290A	16	97.96
^{244}Pu	297C	20	87.69
^{240}Pu	HIP1068	21, 22	99.86
^{239}Pu	453BO	23	99.10
^{241}Pu	307A	24	96.77
^{232}Th	4151	25	100.00
^{236}U	201DMR	26	88.96
^{234}U	M9	27	99.764
^{235}U	264C	28	99.89
^{231}Pa	PaF1	29	100.00
^{238}Pu	06HP014	30	99.38
^{230}Th	256A	31	89.39
^{233}U	240A	32	99.886

Dosimeter materials were selected to help determine neutron fluxes; three positions were chosen to help determine axial fluctuations in flux level. Figure 1 shows the sample locations for dosimeters in the FPs; Table 2 shows the materials and their enrichments. Additional details are given in Refs. 1 and 2.

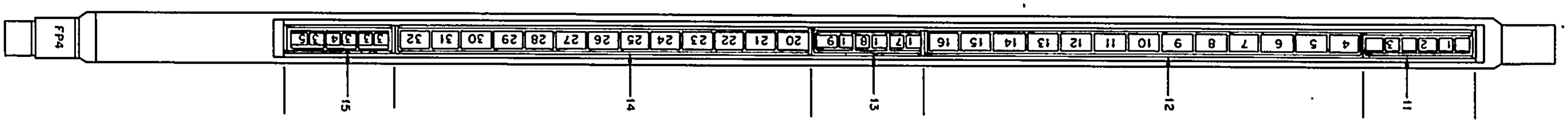

Fig. 1. Sample locations for physics specimens and dosimeter capsules in fuel pins.

Table 2. Dosimeter materials selected for irradiation

Isotope	Batch No.	Position in FP	Isotope enrichment (wt %)
^{235}U	264C	1, 17, 33	99.89
^{238}U	ESZ	1, 17, 33	99.999
^{239}Pu	453BO	2, 18, 34	99.10
^{237}Np	24HP	2, 18, 34	99.99
Cu	N	3, 19, 35	Normal
Co	Co-1	3, 19, 35	Normal

3. METHODS USED FOR ACTINIDE ANALYSIS

3.1 INTRODUCTION

A combination of high-sensitivity mass spectrometry and radiometric methods was used to analyze the various actinides and neutron reaction products. Milligram quantities or less of materials made accurate analyses very difficult. The removal and identification of each capsule without loss or damage were two of many very difficult operations, especially because these tasks must be accomplished in a hot cell. Another very difficult and time-consuming step was the dissolution of the materials in the hot cells after the materials were removed from the FPs. The dissolution methodology for the various materials is discussed in Sect. 3.2.

3.2 DISSOLUTION OF ACTINIDE PHYSICS SPECIMENS AND DOSIMETER MATERIALS

3.2.1 Arrangement of FP Assembly

For the U.S.–U.K. actinide experiment, FPs irradiated in the Dounreay prototype fast reactor (PFR), were comprised of five separate stainless steel tubes (see Fig. 1) in which various vanadium-encapsulated actinide materials were packaged as described in Ref. 1. Three of the tubes contained dosimeter material (six samples per tube) for measurement of the neutron flux to which the assembly was exposed. The remaining two tubes contained the actinide physics specimens. These were packaged 13 samples per tube (see Fig. 1). The tubes then were placed in a stainless steel jacket and arranged such that each physics specimen tube was bracketed on each end by a dosimeter package. All the stainless steel tubes were crimped at both ends to prevent movement of the individual specimens within them.

3.2.2 Removal and Identification of Specimens and Dosimeters

Prior to shipment to the hot-cell facility (where the isolation and dissolution of the individual specimens were performed) the inner stainless steel jacket was removed from the FP and cut into two sections to facilitate shipping. Individual specimens were approximately 1 cm in length; a unique dot code was imprinted on the side near the base of each capsule (see Ref. 2). The physics specimens from the opened tube were removed by simply pouring them out. The remaining four tubes were removed from the jacket by cutting portions of the jacket away until the tubes were exposed; then the tubes were pulled out with pliers. The individual specimens were removed from the stainless steel tubes by mechanically opening both ends of the tube and then pushing the specimens through the opening with a rod. Some difficulty was encountered at this stage because of swelling of the individual specimens.

An in-cell Kollmorgen periscope at 20X magnification was used to examine each capsule and to identify the capsules by the dot codes imprinted on them. All 35 capsules were recovered from the FP without visual damage. After each capsule had been visually examined and identified, it was stored in a clean glass bottle.

3.2.3 Reagents and Glassware

Nitric acid, hydrofluoric acid, and hydrochloric acid used during the dissolution steps were Ultrex-grade (equivalent to double distilled). Water used for dilutions was obtained by passing distilled water through an ion-exchange column filled with mixed anion and cation resins and then by redistilling this product with a quartz still. New glassware (quartz for steps involving hydrofluoric acid) was used for each dissolution. Prior to use, all glassware was soaked overnight in 8 *N* nitric acid. Upon removal from the acid bath, the glassware was rinsed first with distilled water and then a high-purity water and allowed to dry. Plastic reagent bottles were rinsed with nitric acid, distilled water, high-purity water, and reagent-grade ethanol and allowed to dry prior to use.

3.2.4 Summary of Techniques Used for Dissolution of Physics Specimens

All dissolutions of the actinide specimens were performed in a radiochemical hot cell. Dissolutions were performed in a 50-mL quartz beaker equipped with a quartz watchglass and quartz boiling chips. Each capsule was rinsed with acetone before dissolution was started to remove any external contamination. Dissolution of the individual capsules was begun by adding 5 mL of 8 *N* nitric acid to dissolve the vanadium encapsulation material. No heat was used during this step. Dissolution of the vanadium began immediately, as was evidenced by the generation of gas bubbles, and was completed within 1 h. At this point, the solution was blue, and the actinide material rested on the bottom of the beaker in a single rod-shaped segment.

After dissolution of the vanadium, the majority of the solution was transferred to another beaker while the actinide material was undergoing dissolution. Heat was provided to the solution by a hotplate and kept at a temperature at which vapor condensed on the watchglass—but below the solution boiling point. Additional acid was added to the quartz beaker during the dissolution, as needed, to maintain a solution volume of about 10 to 15 mL. The time required to complete the dissolution of a sample varied from 2 h to 2 weeks. Dissolution was termed complete if no solid material could be observed when the beaker was placed on a white background and viewed with a telescope and mirror arrangement.

After the actinide material was dissolved, the vanadium-containing solution was transferred back to the beaker containing the dissolved actinide, and the total volume was reduced to about 10 to 15 mL. The solution was cooled, transferred quantitatively to a tared 25-mL volumetric flask, brought to 25 mL with 8 *N* nitric acid, and weighed. For long-term storage, the solutions were transferred to acid-leached, glass, screw-capped bottles.

Dissolution of the uranium and curium specimens was relatively easy. It was completed in a period ranging from several hours to several days. Only nitric acid and heat were used to dissolve the material.

Dissolution of americium, plutonium, and thorium specimens was difficult, taking up to 2 weeks to put some specimens in solution. Nitric acid, hydrofluoric acid, and heat were used to dissolve the actinide material. Hydrochloric acid was added to several of the plutonium and thorium dissolutions in an attempt to increase the speed of dissolution. However, the addition of hydrofluoric acid produced no observable improvement in the dissolution rate.

Dissolution of neptunium and protactinium specimens from FP-1 and FP-2 took several days. To dissolve the FP-4 specimens in less time, they were subjected to a mixture of nitric acid and hydrofluoric acid on removal of the vanadium-containing solution. This procedure resulted in complete dissolution of the samples with no problems.

3.2.5 Problems Causing Loss of Sample

Leaving the vanadium-containing solution in the original beaker presented problems when the solution volume reached a level low enough such that a vanadium precipitate formed. In the presence of this precipitate, the solution would "bump" vigorously, resulting in the loss of a portion of the sample. Several samples were compromised because of this phenomenon. A second problem with bumping occurred when the dissolution lasted more than a few days. In these samples the sudden generation of a single, large air bubble would occur when the samples became too hot. Even with quartz boiling chips present, the bumping that occurred resulted in the loss of portions of at least four samples. The addition of fresh boiling chips to the beaker did not alleviate the problem. The control of the amount of heat provided to the solution was critical to the effectiveness of the dissolutions and required constant monitoring by a technician. One sample was lost when the beaker in which it was being dissolved was inadvertently overturned.

3.2.6 Dissolution of Dosimeters

The dissolution of the dosimeter specimens, especially from FP-4, presented a unique problem. Within each vanadium capsule were two individual samples which, preferably, would be separated prior to dissolution. Because of the small size of the vanadium pin, it was necessary to remove the sample from the hot cell prior to cutting the sample open, because the manipulator arms used in the hot cell do not provide the dexterity required for this delicate operation. With the ends of the capsule removed, the two samples from FP-1 and FP-2 were pushed out and separated. Dissolution was performed in a radiochemical hood similar to that used on the physics specimens. However, the dosimeter specimens from FP-4 gave radiation readings (i.e., beta/gamma) above that allowed for glove-box operation. It is for this requirement that the dosimeter pairs were dissolved together (see Appendix C).

3.3 DESCRIPTION OF MASS SPECTROMETER USED FOR ACTINIDES ANALYSIS

The limited quantities of the samples for irradiation required that sensitive methods be used for measurements. A combination of high-sensitivity mass spectrometry and radiometric methods generally met these requirements (see Appendix A).

The mass spectrometer used in this work is of the "KAPL" configuration consisting of double deflection magnets. Described in a Knolls Atomic Power Laboratory paper,[6] it possesses ion-counting capability for isotopic composition and isotope dilution measurements. The ions are magnetically deflected through 90° on a 30-cm radius. Ions are detected with an electron multiplier (currently using Hamamatsu Type R515), amplified, and stored on a disk as pulses. The stored information is transferred to a Micro-VAX system for calculation and data processing.[7] A slit midway between the magnets allows only a single mass to be admitted to the second stage. This process results in a spectrum that at high mass is very clean and free from scattered ions. The magnetic field is about 0.8 tesla across a gap of 14.5 mm. The accelerating voltage for this field and the mass region for the actinides is approximately 8 kV. The vacuum system is all metal except for the glass that covers the source housing. The source region is evacuated using a cryogenic pump; the rest of the system (isolated by a valve) is evacuated with ion pumps. Figures 2 and 3 show the mass spectrometer and its associated electronics.

The preparation of samples for loading into the instrument is accomplished using the resin-bead technique.[8,9] For elements that adsorb on anion resin (e.g., Th, Pu, and U), a solution of the element, adjusted to 8 *M* in HNO_3 is contacted with about ten resin beads. These are washed with 8 *M* HNO_3, and a single resin bead is isolated and transferred to the single, zone-refined rhenium filament for isotopic analysis. Trivalent elements that do not adsorb on anion resin under these conditions are purified by solvent extraction and loaded from a weak acid solution onto cation beads (these include americium and curium). The resin-bead loading technique has several advantages: (1) better control of radioactivity in the mass spectrometer and sample introduction area; (2) improved ion optics from the point source; and (3) greater ionization efficiency that allows for more stable ion signals and, hence, better precision of measurement.

For any actinide, spectrum scanning and data recording consist of taking the horizontal sweep voltage from a sweep controller and applying it in series with the accelerating voltage. The sweep voltage is a stair-step function that successively gates the memory channels of the sweep controller. The voltage is thus swept in synchronism with the channel gating so that counts occurring at a given voltage are stored in the same channel during each sweep across the mass spectrum. Any mass in the spectrum can be scanned a predetermined number of times for each traversal of the entire mass range. This procedure allows the scanning scheme to be optimized for the requirements of a given actinide.

For any particular analysis, the counting rate for the major isotope is kept, if possible, at 2×10^5 to 3×10^5 counts/s to avoid a high "count loss" correction. A bias correction is also required because the accelerating voltage is swept, and fractionation effects are inherent in thermal ionization methods. The count loss or system dead-time correction and bias correction are determined by measuring the composition of National Institute of Standards and Technology SRM-U500 standard reference material (SRM). From the observed ratios the bias correction required per mass unit and the system dead time are calculated. Because the elements being measured do not differ drastically in mass from uranium, the corrections are assumed to apply. After the corrections are established, analyses of standards are carried out for quality control purposes.

The system is checked for instrument stability and calibration by running controls using either uranium and/or plutonium certified isotopic standards. These controls are analyzed under the same conditions as are samples. All are on resin beads. Isotopic results given in Table 3 are typical for a mixture of SRM-947 and SRM-500 standards. These results indicate that precisions of ±0.5% (for minor isotopes) and ±0.1% (for major isotopes) are obtained in the 1% and 50 to 99% concentration ranges, respectively.

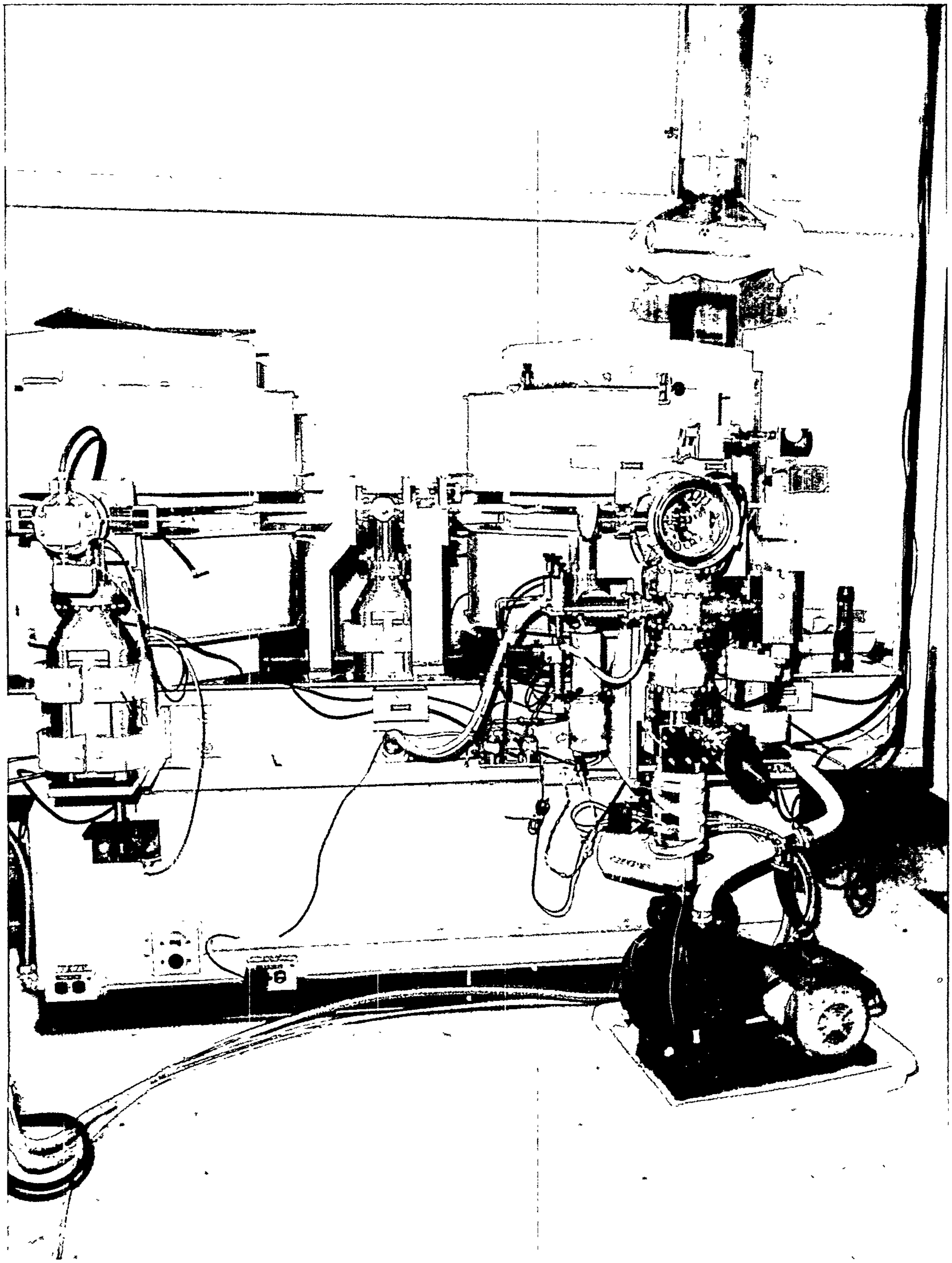

Figure 2. Mass spectrometer for analysis of actinides

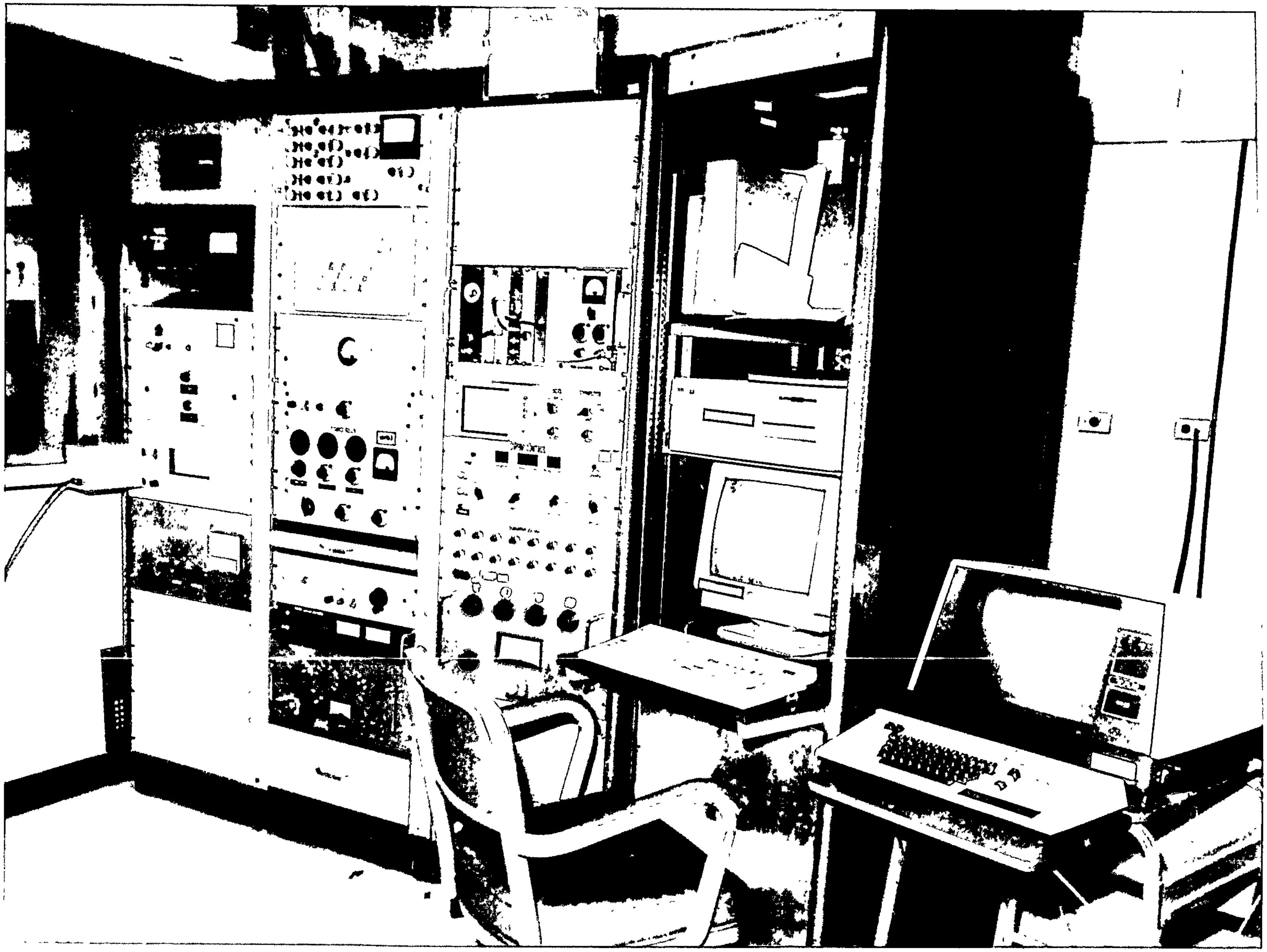

Figure 3. Electronic console for mass spectrometer

Table 3. Mixed plutonium and uranium standards on resin beads[a]

	SRM-947 plutonium			
	238/239	240/239	241/239	242/239
NIST[b]	0.00370	0.24147	0.04309	0.01559
ORNL[c]	0.00371	0.24156	0.04281	0.01559
Std. dev.	±0.00002	±0.00057	±0.00025	±0.00008

	SRM-500 uranium		
	234/235	235/238	236/235
NIST	0.01042	0.99971	0.001519
ORNL	0.01034	0.99851	0.001522
Std. dev.	±0.00005	±0.00209	±0.000005

[a] Nine analyses run over a 2-month period.
[b] National Institute of Standards and Technology.
[c] Oak Ridge National Laboratory.

3.4 DESCRIPTION OF RADIOMETRIC METHODS USED FOR ACTINIDE ANALYSIS

3.4.1 Plutonium-238

Plutonium-238, with a half-life of 87 years, comprises a very small fraction of the material in these samples and cannot usually be analyzed by mass spectrometry because of the interference of ^{238}U. Aliquots of the solution from the dissolution described in Sect. 3.2.4 are diluted to 1 *M* HNO_3, and the plutonium valence is adjusted to the tetravalent state with hydroxylamine hydrochloride and sodium nitrite. Residual fluoride is complexed with aluminum. Pu^{4+} is extracted into 0.5 *M* thenoyltrifluoroacetone (TTA) in xylene. For counting the alpha activity on a gas-flow proportional (2π geometry) counter, aliquots of the organic phase are evaporated onto polished stainless steel planchets. The alpha pulse-height distribution on these plates is measured on a surface barrier detector coupled with a multichannel analyzer. Enough counts are collected to give at least 1000 counts in the 5.49-MeV peak of ^{238}Pu. The total alpha count, percent of ^{238}Pu alpha, sample weight, and dilution factor are used to calculate the ^{238}Pu concentration.

3.4.2 Americium and Curium

Aliquots of the solution from the dissolution (or of appropriate dilutions) are evaporated onto stainless steel planchets, and the gross alpha activity is measured by gas-flow proportional counting. Alpha pulse-height counting gives the ratios of ^{243}Am (5.27 MeV), ^{241}Am (5.48 MeV), and ^{244}Cm (5.81 MeV). The amount of each nuclide can be calculated from the gross alpha activity and their respective specific activities. Measurement of the 60-

keV gamma from ^{241}Am using a Ge(Li) detector coupled with a multichannel analyzer gives a further check on the ^{241}Am content.

3.4.3 Neptunium-237

Aliquots of the dissolved sample are spiked with known amounts of ^{239}Np tracer and are treated with $K_2Cr_2O_7$ to oxidize neptunium (and plutonium) to the hexavalent state. The sample is adjusted to 2 *M* $Al(NO_3)_3$, and the neptunium is extracted (along with plutonium and uranium) into hexone. The neptunium and plutonium are stripped from the organic phase into a reducing solution containing $FeCl_2$ in 1 *M* HCl. The reduced neptunium is then extracted into 0.5 *M* TTA, while the Pu^{3+} remains in the aqueous phase. Aliquots of the organic phase are plated and counted for alpha activity as previously described. Alpha pulse-height analysis is again used to determine the amount of 4.78-MeV ^{237}Np as well as any plutonium carryover. To determine the efficiency of neptunium recovery, an aliquot of the TTA phase is counted for ^{239}Np gamma. The alpha activity of ^{237}Np is corrected for the recovery.

3.4.4 Protactinium-231

Samples for protactinium analysis must be kept in at least 4 *M* HCL solutions. An aliquot of this solution is adjusted to 6 *M* HCl and contacted with di-isobutyl carbinol (DIBC) to extract the protactinium. The organic phase is scrubbed with 6 *M* HCl, and aliquots of the DIBC are evaporated onto tantalum planchets and counted for alpha activity. Purity of the protactinium fraction (5.01 MeV) is checked by pulse-height analysis. Because the gamma tracer, ^{233}Pa, is not readily available, recovery estimates are based on analysis of protactinium solutions of known concentration.

3.4.5 Uranium-232

Uranium is separated from other radioelements by anion exchange. Uranium in 6 *M* HCl is adsorbed on the resin and washed with HCl to remove Th, Am, Cm, Pu, and Np. Elution with dilute (0.01 *M*) HCl gives a purified uranium solution for alpha pulse-height analysis. The 5.32-MeV ^{232}U and 4.82-MeV ^{233}U alpha peaks are integrated, and the ratio of ^{232}U to ^{233}U is determined.

4. RESULTS AND DISCUSSION OF ACTINIDE ANALYSIS

Appendixes B and C contain sample data for each target isotope material that is listed at the top of each page. The information given for each target isotope is broken down into three parts: (A) Part A contains the isotopic composition (in atoms %) of the element with the highest available isotope enrichment of the target isotope as described in Refs. 1 and 2. Isotopic analysis and dates of measurement are shown for FP-1, FP-2, and FP-4 after irradiation under separate column headings. (B) Part B indicates the estimated weight of the target isotope that was encapsulated in vanadium before its fabrication into pins. The measured weight of the target isotope after irradiation and the dates for measurement are given under the appropriate headings for FP-1, FP-2, and FP-4 when reading from top to bottom of the page. (C) Part C contains isotopic and concentration data for important nuclear reaction products when present in sufficient quantity to be measured by currently available techniques.

Results for concentration have been obtained using isotope dilution mass spectrometry and radiochemical counting methods. Isotopic composition results are generally of high quality and were shown to be very reproducible when replicates were analyzed. Although agreement between measured and expected concentration values in the majority of results appears good, the difference in some is much greater than would be expected and lies outside the proven performance of the methods employed. The difference and large variance are disturbing for some results and could be caused by errors resulting from conditions not easily controlled (e.g., target loading and encapsulation, starting material stoichiometry, weighing, dissolution, contamination in hot cells or glove boxes, and aliquoting). Error bars have not been assigned to results because of the uncertainties. However, overall errors on concentration values are estimated between 3% and 5% when optimum measurable quantities could be taken. These would include isotopes of U, Pu, Am, and Th. Other more difficult analyses (involving counting methods where isotopes of interest were minor contributors of the total counts because of differing half-lives) could be performed only on a best-effort basis. These may include isotopes of Am, Np, Cm, Pa, and Pu.

We have made a preliminary comparison of experimental results with calculations using the most recently available cross-section data (ENDF/B-VI). A total of 26 samples for 21 actinides were present in FP-4 (see Table 1). Sample No. 8 (^{244}Cm) was lost during dissolution. We had direct evidence that substantial portions of samples No. 32 (^{233}U), No. 11 (^{238}U), and No. 23 (^{239}Pu) were lost during dissolution. For 18 of the remaining 22 samples (14 actinides), the calculated concentrations of the principal actinides were within 15% of the measured values. This comparison also showed that some sample loss probably occurred also for samples No. 29 (^{231}Pa), No. 26 (^{236}U), No. 30 (^{238}Pu), and No. 16 (^{242}Pu). These uncertainties clearly outweigh all other uncertainties in the measurements.

5. GAMMA-RAY ASSAY MEASUREMENTS OF FP-4 SAMPLES FOR FISSION PRODUCT YIELDS

In this section we discuss the experimental details for FP-4; these are quite similar to our previous reports for the samples in FP-1[4] and FP-2.[5]

Preparation of the actinide samples for the gamma-ray assay measurements followed standard chemical procedures as described in Sect. 3.2, and an aliquot from each actinide sample was extracted for sample preparation. For 12 of the actinide samples, the extracted aliquot was dried on a flat surface of 0.02-mm-thick clear plastic affixed to a 0.9-mm-thick aluminum plate having a 2.54-cm-diam centered hole. The aliquot was dried and then covered with a similar clear plastic to fix the sample material in the center hole. For these samples, then, unlike the liquid samples of FP-1[4] and FP-2,[5] corrections for gamma-ray attenuation by the liquid and bottle were eliminated. However, the aliquot size was determined by the alpha activity of the aliquot, which, in turn, determined the aliquot obtained for these measurements. For the remaining actinides, the alpha intensity was too large for fabrication as a dried sample with a sufficient aliquot to do the gamma-ray assay measurements. For these 13 samples, the aliquot amount was determined by requiring that the dose for a given liquid sample be less than 40 mR/h. The bottles used were smaller than those used for FP-1 and FP-2 samples, and the aliquot for each sample was diluted to a volume of 0.5 cc. For these samples, a special sample holder was fabricated to hold the sample upright and at a predetermined distance from the gamma-ray detector. The given initial sample (isotopic) masses,[2] the present aliquots, and the sample configuration (dried solid or liquid) are tabulated in Table 4.

In addition to the actinide samples, 11 samples containing aliquots of dosimeters were obtained and counted. These included separated samples for the cobalt-copper dosimeters (six samples in all) assayed for ^{60}Co, three samples that combined the ^{237}Np and ^{239}Pu dosimeters assayed for ^{137}Cs, and two (of the original three) samples that combined the ^{235}U and ^{238}U dosimeters also assayed for ^{137}Cs.

The aliquots were counted using two germanium detector systems, one a 15% efficiency Ge(Li) detector having a resolution full width at half maximum (FWHM) of ~2.1 keV for $E_\gamma = 1.33$ MeV. This one was used for measurement of gamma rays having energies between 0.1 and 3 MeV, and the other, a small planar detector having a resolution FWHM of ~0.5 keV for $E_\gamma = 122$ keV, was used for measurement of gamma rays having energies between 10 and 400 keV. The latter detector also detected beta rays emanating from the dried samples; thus some of the measurements with these samples included a 3.2-g/cc carbon absorber placed between the sample and the detector. Gamma-ray attenuation by the carbon absorber was corrected using the absorption tables of Storm and Israel.[10] For the liquid samples, attenuation through the bottom of the glass bottle and by the liquid was also computed.

For each of the actinide samples, at least one spectrum was obtained using each detector. Measurements were initiated in March 1990 (about 600 days following the end of the irradiation of the fuel pin) and essentially were completed in May 1990; two additional spectra were obtained in 1992 to help clarify ambiguous results obtained during the 1990 measurements. Data reduction was accomplished using the documented code TPASS[11] combined with manual methods when required. Computed peak yields were corrected for detector efficiency and (if needed) absorption; peak energies were then compared with

Table 4. FP-4 principal actinide masses

Principal isotope	Sample No.	Total isotopic mass (mg)[a]	γ-ray assay aliquot (%)	Sample status
^{230}Th	31	2.883	1.00	Solid
^{232}Th	25	18.880	1.00	Solid
^{231}Pa	29	2.512	0.10	Solid
^{233}U	32	7.652	0.30	Solid
^{234}U	27	3.500	1.00	Solid
^{235}U	28	8.373	0.40	Solid
^{236}U	26	7.962	1.00	Solid
^{238}U	11	9.810	2.00	Solid
^{237}Np	7	12.216	1.00	Solid
^{238}Pu	30	2.889[b]	2.00	Liquid
^{239}Pu	23	8.470	0.30	Liquid
^{240}Pu	21	10.839	0.60	Liquid
^{240}Pu	22	10.816	0.80	Liquid
^{241}Pu	24	4.136[b]	0.40	Liquid
^{242}Pu	16	2.046	2.00	Solid
^{244}Pu	20	2.108	2.00	Solid
^{241}Am	14	9.422	0.20	Liquid
^{241}Am	15	9.525	0.40	Liquid
^{243}Am	12	9.831	0.20	Liquid
^{243}Am	13	9.801	0.40	Liquid
^{243}Cm	10	0.373[b]	0.10	Liquid
^{244}Cm	9	7.940[b]	0.60	Liquid
^{246}Cm	5	6.691	0.20	Liquid
^{246}Cm	6	6.797	1.00	Liquid
^{248}Cm	4	1.765	1.00	Solid

[a] From Table 5 of Ref. 2
[b] Recomputed as of July 17, 1982.

nuclear data for specific radionuclides to determine the number of atoms in the aliquot as of the end of the irradiation (EOI), which was taken to be July 7, 1988. The nuclear data[12,13] for the fission products studied in this analysis are given in Table 5. Uncertainties associated with half-lives were not propagated in the computations; however, uncertainties in branching ratios were included in the final uncertainty determinations. Particular interest was centered on the production of the "shielded" radionuclides ^{110m}Ag, ^{134}Cs, ^{152}Eu, and ^{154}Eu. Other longer-lived fission product yields were also determined. Results for ^{137}Cs are given in Table 6; these results are compared with similar data deduced for the measurements of FP-2. To zero-order, the ratios of the ^{137}Cs yields for the actinides of FP-4 to those for FP-2 ought to be ~8 based on the ratio of total irradiation of the FP-4 in the PFR with respect to the FP-2. The observed variations give some indication of the effects of the long irradiation (e.g., the "growing in" of ^{233}U in the ^{232}Th sample by capture, which increases the ^{137}Cs production because ^{233}U has a much larger fission cross section than does ^{232}Th). Yields for four other longer-lived fission products are given in Table 7, and yields for the "shielded" radionuclides are presented in Table 8.

In addition, the data were studied to obtain yields of long-lived actinides in the samples. Of particular interest, based on the FP-1 experience, were the yields of principal actinides that could be deduced directly from gamma-ray assay. These are presented in Table 9 in the fourth column. For comparison, values estimated from the radiometric results of Sect. 3.4 are given in the final column. These were deduced from data given in Appendix B as follows: in the table for ^{238}Pu, capsule position 30, in the last column for (FP-4) the concentration after irradiation is reported as 0.65 mg as of March 26, 1990. The γ-ray assay aliquot for this sample was 2.0%. Therefore, the radiometric ^{238}Pu concentration to be compared with the γ-ray assay concentration should be 2% of 0.65 mg, or 13 μg. Similar values for the other six samples in Table 9 were also deduced. A 5% uncertainty to these masses was assigned based on the discussion in Sect. 4. The γ-ray assay measurements tend to be smaller than the radiometric estimates by ~12% on the average. Two potential sources of error in the gamma-ray assay are (a) the exact determination of the masses of the small aliquots and (b) the exact determination of the detection efficiencies including photon attenuations by the glass bottles. The relative actinide mass yields are excellent, and an uncertainty of 10% assigned to these data would be reasonable.

Actinides produced by means of neutron capture by the principal actinides, or else the beta-decay products of the radionuclides produced by capture, are given in Table 10. Finally, in Table 11 are given yields of other actinides observed and delineated in the various spectra. Some of these actinides (e.g., ^{228}Th) were likely present in some of the samples before the irradiation started, while others were created by neutron interactions with nonprincipal (or minor) actinides in the samples. Lastly, the results of the dosimeter studies are given in Table 12.

Table 5. Nuclear data for fission products

Radionuclide	Half-life	Gamma ray	
		Energy (keV)	Branching (%)
^{106}Rh[a]	366.9 d	621.8 1050.1	9.95 ± 0.75 1.45 ± 0.10
^{110m}Ag[b]	252. d	884.2 937.4 1383.9	72.6 ± 0.3 34.2 ± 0.3 24.3 ± 0.2
^{125}Sb	2.758 y	427.89[c] 600.56 635.90 463.38	29.44 ± 0.99 17.78 ± 0.95 11.32 ± 0.68 10.45 ± 0.57
^{134}Cs	2.066 y	604.74 795.80	98.6 ± 0.3 87.8 ± 1.4
^{137}Cs	30.14 y	661.64[d]	86.0 ± 0.6[d]
^{144}Ce	284.7 d	133.53	11.09 ± 0.20
^{144}Pr[e]	284.7 d	696.48 2185.78	1.34 ± 0.02 0.74 ± 0.03
^{152}Eu	13.6 y	1407.92	20.7 ± 0.7
^{154}Eu	8.75 y	1274.8 1004.8	33.6 ± 0.7 17.6 ± 0.4
^{155}Eu	4.96 y	86.55 105.31	30.9 ± 2.8 20.7 ± 2.0

[a] Listed half-life is that of the parent ^{106}Ru.

[b] The peak corresponding to detection of the E_γ = 657.7-keV gamma ray from decay of this isotope is masked by the very much larger peak at 661.6 keV corresponding to decay of ^{137}Cs.

[c] Nearly degenerate with E_γ = 428.4 keV due to decay of ^{106}Rh.

[d] Gamma ray due to decay of daughter ^{137}Ba isomer; branching corrected to be applicable to decay of ^{137}Cs.

[e] Listed half-life is that of the parent ^{144}Ce.

Table 6. Absolute experimental yields (in the aliquots) of the fission product ^{137}Cs as of the end of the PFR irradiation[a]

Principal isotope	Sample No.	Yields (atoms)		Ratio FP-4:FP-2
		FP-4	FP-2[b]	
^{230}Th	31	$(8.51 \pm 0.07) \times 10^{14}$	1.98×10^{13}	43.0
^{232}Th	25	$(7.96 \pm 0.44) \times 10^{14}$	1.11×10^{13}	71.8
^{231}Pa	29	$(4.78 \pm 0.27) \times 10^{13}$	1.49×10^{12}	32.0
^{233}U	32	$(1.77 \pm 0.05) \times 10^{15}$	1.602×10^{14}	11.1
^{234}U	27	$(5.83 \pm 0.33) \times 10^{14}$	3.00×10^{13}	19.5
^{235}U	28	$(9.21 \pm 0.26) \times 10^{14}$	1.64×10^{14}	5.6
^{236}U	26	$(6.74 \pm 0.38) \times 10^{14}$	6.33×10^{13}	10.6
^{238}U	11	$(2.31 \pm 0.06) \times 10^{14}$	2.97×10^{13}	7.8
^{237}Np	7	$(1.77 \pm 0.10) \times 10^{15}$	6.94×10^{13}	25.5
^{238}Pu	30	$(7.40 \pm 0.48) \times 10^{14}$	6.84×10^{13}	10.8
^{239}Pu	23	$(1.12 \pm 0.07) \times 10^{15}$	1.39×10^{14}	8.0
^{240}Pu	21	$(1.14 \pm 0.07) \times 10^{15}$	8.10×10^{13}	14.1
^{240}Pu	22	$(1.49 \pm 0.10) \times 10^{15}$	1.08×10^{14}	13.8
^{241}Pu	24	$(8.30 \pm 0.54) \times 10^{14}$	9.45×10^{13}	8.8
^{242}Pu	16	$(1.44 \pm 0.04) \times 10^{14}$	2.53×10^{13}	5.7
^{244}Pu	20	$(4.95 \pm 0.28) \times 10^{14}$	3.53×10^{13}	14.0
^{241}Am	14	$(2.61 \pm 0.29) \times 10^{14}$	1.80×10^{13}	14.5
^{241}Am	15	$(5.23 \pm 0.34) \times 10^{14}$	3.60×10^{13}	14.5
^{243}Am	12	$(1.48 \pm 0.05) \times 10^{14}$	1.26×10^{13}	11.7
^{243}Am	13	$(2.93 \pm 0.08) \times 10^{14}$	2.41×10^{13}	12.2
^{243}Cm	10	$(4.13 \pm 0.11) \times 10^{13}$	5.80×10^{12}	7.1
^{244}Cm	9	$(8.52 \pm 0.55) \times 10^{14}$	8.01×10^{13}	10.6
^{246}Cm	5	$(1.76 \pm 0.11) \times 10^{14}$	2.26×10^{13}	7.8
^{246}Cm	6	$(8.81 \pm 0.57) \times 10^{14}$	1.15×10^{14}	7.7
^{248}Cm	4	$(1.64 \pm 0.05) \times 10^{14}$	1.78×10^{13}(c)	9.2

[a] Sept. 1, 1983 for FP-2; July 7, 1988 for FP-4.

[b] Renormalized from data in Table 10 of Ref. 5 to the same computed masses used in the present experiment.

[c] Datum from FP-1 (Ref. 4).

Table 7. FP-4: yields of fission products deduced by gamma-ray assay

Principal isotope	Sample No.	Fission-product yield (atoms)[a]			
		^{106}Ru[b]	^{125}Sb	^{144}Ce[c]	^{155}Eu
^{230}Th	31		$(8.40 \pm 0.33) \times 10^{11}$	$(3.33 \pm 0.06) \times 10^{13}$	$(1.34 \pm 0.13) \times 10^{11}$
^{232}Th	25	$(2.82 \pm 0.20) \times 10^{12}$	$(1.26 \pm 0.07) \times 10^{13}$	$(2.88 \pm 0.09) \times 10^{14}$	$(2.33 \pm 0.16) \times 10^{12}$
^{231}Pa	29	$(3.37 \pm 0.35) \times 10^{11}$	$(7.23 \pm 0.40) \times 10^{11}$	$(1.47 \pm 0.15) \times 10^{13}$	$(1.22 \pm 0.09) \times 10^{11}$
^{233}U	32	$(2.10 \pm 0.15) \times 10^{13}$	$(1.89 \pm 0.08) \times 10^{13}$	$(3.45 \pm 0.12) \times 10^{14}$	$(5.22 \pm 0.36) \times 10^{12}$
^{234}U	27	$(1.42 \pm 0.10) \times 10^{13}$	$(4.94 \pm 0.25) \times 10^{12}$	$(1.94 \pm 0.07) \times 10^{14}$	$(2.67 \pm 0.19) \times 10^{12}$
^{235}U	28	$(2.58 \pm 0.18) \times 10^{13}$	$(6.52 \pm 0.22) \times 10^{12}$	$(5.00 \pm 0.15) \times 10^{14}$	$(9.69 \pm 0.69) \times 10^{12}$
^{236}U	26	$(3.28 \pm 0.23) \times 10^{13}$	$(4.26 \pm 0.29) \times 10^{12}$	$(2.18 \pm 0.07) \times 10^{14}$	$(5.80 \pm 0.41) \times 10^{12}$
^{238}U	11	$(4.79 \pm 0.34) \times 10^{13}$	$(3.62 \pm 0.13) \times 10^{12}$	$(6.90 \pm 0.29) \times 10^{13}$	$(5.00 \pm 0.35) \times 10^{12}$
^{237}Np	7	$(5.30 \pm 0.37) \times 10^{13}$	$(2.89 \pm 0.10) \times 10^{13}$	$(4.52 \pm 0.22) \times 10^{14}$	$(3.24 \pm 0.33) \times 10^{13}$
^{238}Pu	30	$(1.28 \pm 0.09) \times 10^{14}$	$(1.91 \pm 0.10) \times 10^{13}$	$(1.43 \pm 0.10) \times 10^{14}$	$(1.16 \pm 0.09) \times 10^{13}$
^{239}Pu	23	$(3.34 \pm 0.24) \times 10^{13}$	$(1.04 \pm 0.08) \times 10^{13}$	$(1.54 \pm 0.05) \times 10^{14}$	$(2.09 \pm 0.14) \times 10^{13}$
^{240}Pu	21	$(2.71 \pm 0.19) \times 10^{13}$	$(1.09 \pm 0.07) \times 10^{13}$	$(3.05 \pm 0.10) \times 10^{14}$	$(3.55 \pm 0.25) \times 10^{13}$
^{240}Pu	22	$(5.47 \pm 0.38) \times 10^{13}$	$(1.40 \pm 0.09) \times 10^{13}$	$(3.63 \pm 0.11) \times 10^{14}$	$(4.27 \pm 0.29) \times 10^{13}$
^{241}Pu	24	$(9.62 \pm 0.68) \times 10^{13}$	$(6.86 \pm 0.42) \times 10^{12}$	$(1.79 \pm 0.06) \times 10^{14}$	$(2.87 \pm 0.20) \times 10^{13}$
^{242}Pu	16	$(1.61 \pm 0.11) \times 10^{13}$	$(1.12 \pm 0.04) \times 10^{12}$	$(4.37 \pm 0.13) \times 10^{13}$	$(5.24 \pm 0.37) \times 10^{12}$
^{244}Pu	20	$(5.05 \pm 0.35) \times 10^{13}$	$(3.59 \pm 0.20) \times 10^{12}$	$(1.45 \pm 0.05) \times 10^{14}$	$(2.78 \pm 0.20) \times 10^{13}$
^{241}Am	14	$(2.90 \pm 0.20) \times 10^{13}$	$(6.47 \pm 0.22) \times 10^{12}$	$(6.46 \pm 0.19) \times 10^{13}$	$(1.04 \pm 0.07) \times 10^{13}$
^{241}Am	15	$(1.14 \pm 0.08) \times 10^{14}$	$(1.35 \pm 0.08) \times 10^{13}$	$(1.35 \pm 0.04) \times 10^{14}$	$(2.11 \pm 0.15) \times 10^{13}$
^{243}Am	12	$(2.06 \pm 0.14) \times 10^{13}$	$(2.83 \pm 0.10) \times 10^{12}$	$(4.04 \pm 0.12) \times 10^{13}$	$(2.57 \pm 0.18) \times 10^{13}$
^{243}Am	13	$(4.12 \pm 0.29) \times 10^{13}$	$(5.63 \pm 0.40) \times 10^{12}$	$(8.12 \pm 0.25) \times 10^{13}$	$(6.13 \pm 0.43) \times 10^{13}$
^{243}Cm	10	$(5.22 \pm 0.36) \times 10^{12}$	$(8.50 \pm 0.29) \times 10^{11}$	$(8.50 \pm 0.26) \times 10^{12}$	$(1.44 \pm 0.10) \times 10^{12}$
^{244}Cm	9	$(3.43 \pm 0.24) \times 10^{14}$	$(1.33 \pm 0.08) \times 10^{13}$	$(2.27 \pm 0.08) \times 10^{14}$	$(5.53 \pm 0.39) \times 10^{13}$
^{246}Cm	5	$(7.94 \pm 0.56) \times 10^{13}$	$(2.35 \pm 0.08) \times 10^{12}$	$(5.30 \pm 0.20) \times 10^{13}$	$(1.42 \pm 0.10) \times 10^{13}$
^{246}Cm	6	$(2.16 \pm 0.15) \times 10^{14}$	$(1.18 \pm 0.09) \times 10^{13}$	$(2.70 \pm 0.16) \times 10^{14}$	$(7.30 \pm 0.51) \times 10^{13}$
^{248}Cm	4	$(5.86 \pm 0.41) \times 10^{13}$	$(2.06 \pm 0.07) \times 10^{12}$	$(5.57 \pm 0.17) \times 10^{13}$	$(1.56 \pm 0.11) \times 10^{13}$

[a] As of July 7, 1988.

[b] From assay of ^{106}Rh.

[c] Includes assay of ^{144}Pr.

Table 8. FP-4: yields of shielded radionuclides deduced by gamma-ray assay

Principal isotope	Sample No.	Radionuclide yield (in atoms)[a]			
		^{110m}Ag	^{134}Cs	^{152}Eu	^{154}Eu
^{230}Th	31		$(2.38 \pm 0.04) \times 10^{12}$		
^{232}Th	25		$(2.34 \pm 0.06) \times 10^{13}$		$(1.63 \pm 0.15) \times 10^{12}$
^{231}Pa	29		$(1.34 \pm 0.05) \times 10^{12}$		$(9.15 \pm 0.50) \times 10^{10}$
^{233}U	32		$(7.52 \pm 0.20) \times 10^{13}$		$(5.29 \pm 0.20) \times 10^{12}$
^{234}U	27		$(2.43 \pm 0.07) \times 10^{13}$		$(2.32 \pm 0.06) \times 10^{12}$
^{235}U	28		$(4.00 \pm 0.10) \times 10^{13}$		$(9.54 \pm 0.33) \times 10^{12}$
^{236}U	26		$(3.25 \pm 0.09) \times 10^{13}$		$(4.35 \pm 0.15) \times 10^{12}$
^{238}U	11	$(5.64 \pm 0.96) \times 10^{10}$	$(8.82 \pm 0.30) \times 10^{12}$		$(2.18 \pm 0.06) \times 10^{12}$
^{237}Np	7	$(2.68 \pm 0.17) \times 10^{11}$	$(6.98 \pm 0.36) \times 10^{13}$		$(1.43 \pm 0.08) \times 10^{13}$
^{238}Pu	30	$(2.27 \pm 0.13) \times 10^{11}$	$(3.59 \pm 0.16) \times 10^{13}$		$(6.40 \pm 0.25) \times 10^{12}$
^{239}Pu	23	$(4.50 \pm 0.21) \times 10^{11}$	$(5.79 \pm 0.26) \times 10^{13}$	$(2.67 \pm 0.17) \times 10^{11}$	$(1.19 \pm 0.06) \times 10^{13}$
^{240}Pu	21	$(7.46 \pm 0.34) \times 10^{11}$	$(4.86 \pm 0.28) \times 10^{13}$	$(1.82 \pm 0.28) \times 10^{11}$	$(1.58 \pm 0.06) \times 10^{13}$
^{240}Pu	22	$(9.51 \pm 0.46) \times 10^{11}$	$(6.44 \pm 0.36) \times 10^{13}$	$(2.43 \pm 0.19) \times 10^{11}$	$(1.90 \pm 0.10) \times 10^{13}$
^{241}Pu	24	$(7.41 \pm 0.32) \times 10^{11}$	$(4.04 \pm 0.22) \times 10^{13}$	$(1.99 \pm 0.23) \times 10^{11}$	$(1.51 \pm 0.09) \times 10^{13}$
^{242}Pu	16	$(1.50 \pm 0.05) \times 10^{11}$	$(6.10 \pm 0.25) \times 10^{12}$	$(2.88 \pm 0.17) \times 10^{11}$	$(3.04 \pm 0.09) \times 10^{12}$
^{244}Pu	20	$(7.74 \pm 0.34) \times 10^{11}$	$(1.87 \pm 0.10) \times 10^{13}$	$(1.32 \pm 0.10) \times 10^{11}$	$(1.08 \pm 0.04) \times 10^{13}$
^{241}Am	14	$(1.47 \pm 0.09) \times 10^{11}$	$(9.81 \pm 0.62) \times 10^{12}$	$(8.24 \pm 0.40) \times 10^{10}$	$(4.55 \pm 0.12) \times 10^{12}$
^{241}Am	15	$(3.90 \pm 0.17) \times 10^{11}$	$(2.02 \pm 0.07) \times 10^{13}$	$(1.61 \pm 0.24) \times 10^{11}$	$(9.21 \pm 0.35) \times 10^{12}$
^{243}Am	12	$(1.56 \pm 0.07) \times 10^{11}$	$(5.20 \pm 0.21) \times 10^{12}$		$(3.39 \pm 0.16) \times 10^{12}$
^{243}Am	13	$(4.01 \pm 0.17) \times 10^{11}$	$(1.04 \pm 0.06) \times 10^{13}$	$(7.35 \pm 0.60) \times 10^{10}$	$(6.75 \pm 0.31) \times 10^{12}$
^{243}Cm	10	$(6.34 \pm 0.33) \times 10^{10}$	$(1.74 \pm 0.07) \times 10^{12}$	$(3.46 \pm 0.30) \times 10^{10}$	$(1.28 \pm 0.04) \times 10^{12}$
^{244}Cm	9	$(1.18 \pm 0.07) \times 10^{12}$	$(3.00 \pm 0.18) \times 10^{13}$	$(3.07 \pm 0.20) \times 10^{11}$	$(1.89 \pm 0.09) \times 10^{13}$
^{246}Cm	5	$(3.10 \pm 0.15) \times 10^{11}$	$(5.83 \pm 0.34) \times 10^{12}$	$(6.72 \pm 0.50) \times 10^{10}$	$(4.72 \pm 0.22) \times 10^{12}$
^{246}Cm	6	$(7.32 \pm 0.43) \times 10^{11}$	$(2.96 \pm 0.13) \times 10^{13}$	$(2.53 \pm 0.16) \times 10^{11}$	$(2.40 \pm 0.11) \times 10^{13}$
^{248}Cm	4	$(3.40 \pm 0.14) \times 10^{11}$	$(5.54 \pm 0.23) \times 10^{12}$		

[a] As of July 7, 1988.

Table 9. FP-4: comparison of post-irradiation actinide sample masses measured by two methods. The listed values are for the aliquots.

Principal isotope	Sample No.	Aliquot[a] (%)	Mass (μg)	
			Gamma-ray assay	Radiometric
^{238}Pu	30	2.0	11.7 ± 0.5	13 ± 1
^{241}Pu	24	0.4	7.5 ± 0.5	7.8 ± 0.4
^{241}Am	14	0.2	11.1 ± 0.5	12.4 ± 0.6
^{241}Am	15	0.4	21.7 ± 1.1	25.4 ± 1.3
^{243}Am	12	0.2	11.7 ± 0.4	13.6 ± 0.7
^{243}Am	13	0.4	23.2 ± 0.8	26.4 ± 1.3
^{243}Cm	10	0.1	0.190 ± 0.010	0.22 ± 0.01

[a] From Table 4.

[b] At the end of irradiation (July 7, 1988).

[c] From tabular value in Appendix B, row B "After irradiation," final column multiplied by the γ-ray aliquot at date given in the next row. Uncertainty assumed to be 5%.

Table 10. FP-4: actinides produced in neutron capture by principal isotopes deduced by gamma-ray assay. The listed values are for the aliquots.

Principal isotope	Sample No.	Aliquot[a] (%)	Capture product	Yield[b,c] (atoms)
^{231}Pa	29	0.1	^{232}U	1.50×10^{15}
^{240}Pu	21	0.6	^{241}Am	2.24×10^{15} at EOI + 651 d
^{240}Pu	22	0.8	^{241}Am	2.71×10^{15} at EOI + 664 d
^{242}Pu	16	2.0	^{243}Am	2.81×10^{15}
^{244}Pu	20	2.0	^{245}Cm	4.33×10^{15}
^{241}Am	14	0.2	^{242m}Am[d]	4.70×10^{15}
^{241}Am	15	0.4	^{242m}Am[d]	1.00×10^{16}
^{244}Cm	9	0.6	^{245}Cm	8.07×10^{15}
^{248}Cm	4	1.0	^{249}Cf	1.70×10^{15}

[a] From Table 4.

[b] At end of irradiation (July 7, 1988), except as noted.

[c] Uncertainty is ±5%.

[d] From assay of 16-h ^{242}Am daughter.

Table 11. FP-4: yields of actinides other than principal and capture nuclides as deduced by gamma-ray assay

Principal isotope	Sample No.	Measured actinide	Yield (atoms)	Cooling time[a] (days)
^{230}Th	31	^{228}Th	$(7.41 \pm 0.37) \times 10^{13}$	609
^{230}Th	31	^{232}U	$(1.42 \pm 0.04) \times 10^{15}$	609
^{232}Th	25	^{228}Th	$(1.22 \pm 0.07) \times 10^{12}$	621
^{231}Pa	29	^{228}Th	$(1.06 \pm 0.06) \times 10^{14}$	632
^{233}U	32	^{228}Th	$(1.15 \pm 0.07) \times 10^{11}$	618
^{234}U	27	^{228}Th	$(1.20 \pm 0.07) \times 10^{11}$	629
^{237}Np	7	^{228}Th	$(9.4 \pm 2.0) \times 10^{10}$	609
^{239}Pu	23	^{241}Am	$(2.93 \pm 0.29) \times 10^{13}$	659
^{241}Pu	24	^{241}Am	$(1.51 \pm 0.05) \times 10^{16}$	666
^{242}Pu	16	^{241}Am	$(2.71 \pm 0.24) \times 10^{13}$	679
^{244}Pu	20	^{241}Am	$(3.56 \pm 0.18) \times 10^{14}$	677
^{244}Pu	20	^{243}Am	$(8.88 \pm 0.36) \times 10^{14}$	1355
^{241}Am	14	^{243}Am	$(1.89 \pm 0.09) \times 10^{14}$	1351
^{241}Am	14	^{243}Cm	$(2.45 \pm 0.08) \times 10^{14}$	1351
^{243}Cm	10	^{241}Am	$(4.10 \pm 0.20) \times 10^{13}$	650
^{244}Cm	9	^{241}Am	$(3.57 \pm 0.16) \times 10^{14}$	638
^{246}Cm	5	^{241}Am	$(2.44 \pm 0.24) \times 10^{13}$	649
^{246}Cm	5	^{245}Cm	$(1.86 \pm 0.36) \times 10^{15}$	649
^{246}Cm	5	^{249}Cf	$(2.06 \pm 0.12) \times 10^{14}$	649
^{246}Cm	6	^{245}Cm	$(3.6 \pm 1.3) \times 10^{14}$	665
^{246}Cm	6	^{249}Cf	$(9.60 \pm 0.39) \times 10^{14}$	665
^{248}Cm	4	^{241}Am	$(7.8 \pm 1.2) \times 10^{12}$	684

[a] After end of irradiation.

Table 12. FP-4: dosimeter yields deduced by gamma-ray assay

Dosimeter	Position	Mass (mg)	Gamma-ray aliquot (%)	Yield (atoms)[a]	
				^{137}Cs	^{60}Co
^{235}U	1A	1.174	1.0	(5.54 ± 0.17) x 10^{14}	
^{238}U	1B	1.180			
^{239}Pu	2A	1.196	12.0	(7.76 ± 0.29) x 10^{15}	
^{237}Np	2B	1.044			
Co	3A	0.00150	2.0		(4.45 ± 0.11) x 10^{12}
Cu	3B	1.353	2.0		(9.90 ± 0.24) x 10^{11}
^{235}U	17A	1.106	0.4	(2.22 ± 0.07) x 10^{14}	
^{238}U	17B	0.982			
^{239}Pu	18A	1.140	12.0	(8.16 ± 0.34) x 10^{15}	
^{237}Np	18B	1.124			
Co	19A	0.00157	2.0		(5.61 ± 0.14) x 10^{12}
Cu	19B	1.406	2.0		(1.63 ± 0.04) x 10^{12}
^{239}Pu	34A	1.184	12.0	(1.14 ± 0.05) x 10^{16}	
^{237}Np	34B	1.184			
Co	35A	0.00143	2.0		(4.91 ± 0.12) x 10^{12}
Cu	35B	1.433	2.0		(1.01 ± 0.03) x 10^{12}

[a] As of July 7, 1988.

6. CONCLUSIONS

Summarized are the analytical methodologies used to measure the isotopic compositions and concentrations of actinides and their reaction products for FP-1, FP-2, and FP-4 irradiated in the Dounreay prototype fast reactor. These measurements have been made using the best laboratory practices possible under the difficult constraints of the use of highly radioactive materials and the small quantities usually selected or produced in bombardment or fission. Some data from FP-1 and FP-2 are suspect. However, the lessons learned from these were of great value in improving the analytical techniques for the more crucial FP-4.

Similarly, experience gained from the study of gamma-ray assay of the samples from FP-1 and FP-2 was invaluable for measurements involving the samples of FP-4. The much more extensive burnup of FP-4 enhanced the production of "second-order" radionuclides as exemplified by observed yields of shielded fission products as well as the wide variations in observed yields of the radionuclide ^{137}Cs resulting from different fissioning rates between the principal actinide in a sample and "grown-in" actinides.

These data should provide a substantial basis for future analytical analyses of fuel-rod radionuclide composition during and following use of the material in a fast reactor.

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APPENDIX A

METHODS USED TO ANALYZE ACTINIDE MATERIALS

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METHODS USED TO ANALYZE ACTINIDE MATERIALS

The symbols MS and RCA denote mass spectrometry (isotope dilution) and radiochemical analysis (alpha and gamma spectro-scopy), respectively.

Position in FP	Material	Method Used Isotopic by MS	Concentration by MS	Concentration by RCA
1	Dos (U-235)	x	x	
	Dos (U-238)	x	x	
2	Dos (Pu-239)	x	x	
	Dos (Np-237)			x
3	Dos (Cu)			x
	Dos (Co)			x
4	Cm-248	x		x
5	Cm-246	x		x
6	Cm-246	x		x
7	Np-237	x		x
8	Cm-244	x		x
9	Cm-244	x		x
10	Cm-243	x		x
11	U-238	x	x	
12	Am-243	x		x
13	Am-243	x		x
14	Am-241	x		x
15	Am-241	x		x
16	Pu-242	x	x	
17	Dos (U-235)	x	x	
	Dos (U-238)	x	x	
18	Dos (Pu-239)	x	x	
	Dos (Np-237)			x
19	Dos (Cu)			x
	Dos (Co)			x

Appendix A (continued)

		Method Used		
Position in FP	Material	Isotopic by MS	Concentration by MS	Concentration by RCA
20	Pu-244	x	x	
21	Pu-240	x	x	
22	Pu-240	x	x	
23	Pu-239	x	x	
24	Pu-241	x	x	
25	Th-232	x	x	
26	U-236	x	x	
27	U-234	x	x	
28	U-235	x	x	
29	Pa-231			x
30	Pu-238	x		x
31	Th-230	x	x	
32	U-233	x	x	
33	Dos (U-235)	x	x	
	Dos (U-238)	x	x	
34	Dos (Pu-239)	x	x	
	Dos (Np-237)			x
35	Dos (Co)			x
	Dos (Co)			x

Appendix B

ANALYTICAL DATA FOR IRRADIATED ACTINIDE PHYSICS SPECIMENS

(See also Ref. 5 for FP-1 and FP-2 Data)

Sample data for ^{248}Cm, Capsule Position 4

	At start	FP-1	FP-2*	FP-4
A. Isotopic Composition, at. %				
Date:	Feb. 11, 1981	Aug. 13, 1984	Oct. 16, 1986	Oct. 17, 1990
244	3.231	2.84	2.58	2.08
245	0.102	0.149	0.150	0.36
246	4.516	4.54	4.51	4.50
247	0.086	0.137	0.136	0.41
248	92.065	92.33	92.62	92.65
B. Concentration, mg ^{248}Cm				
At start		1.70	1.76	1.76
After irradiation		1.61		1.45
Date of measurement		July 24, 1984		Mar. 26, 1990
C. Additional Postirradiation Data				
		July 24, 1984		Oct. 22, 1991
^{249}Bk wt., mg		0.0036		0.0014

*Portion of sample lost in dissolution; isotopic data are excellent and reproducible (see Ref. 5, p. 38, for duplicate results).

Sample data for ^{246}Cm, Capsule Position 5

A. Isotopic Composition, at. %

Date:	At start Aug 5, 1980	FP-1 Aug. 13, 1984	FP-2* Nov. 17, 1986	FP-4 July 20, 1990
243	0.043	--	--	0.02
244	18.43	16.03	14.86	11.78
245	0.234	0.51	0.52	1.78
246	66.51	67.53	68.63	66.39
247	1.84	2.52	2.54	6.32
248	12.95	13.41	13.45	13.71

B. Concentration, mg ^{246}Cm

At start		6.66	6.81	6.69
After irradiation		6.29	6.10	4.92
Date of measurement		July 24, 1984	April 29, 1987	Mar. 27, 1990

C. Additional Postirradiation Data:

		Sept. 13, 1984	Nov. 17, 1986	June 13, 1990
^{238}Pu, at. %		0.024	0.016	0.062
^{239}Pu, "		0.240	0.182	0.176
^{240}Pu, "		97.40	97.73	92.64
^{241}Pu, "		0.86	0.54	5.24
^{242}Pu, "		1.48	1.53	1.88
^{244}Pu, "		<0.001	<0.001	<0.004
Pu wt., mg		0.27	0.42	0.40

*Duplicate results reported in Ref. 5, p. 39.

Sample data for ^{246}Cm, Capsule Position 6

A. Isotopic Composition, at. %

Date:	At start Aug. 5, 1980	FP-1*	FP-2 Nov. 18, 1986	FP-4 July 20, 1990
243	0.043		--	0.01
244	18.43		15.00	11.80
245	0.234		0.52	1.78
246	66.51		68.20	66.34
247	1.84		2.52	6.34
248	12.95		13.36	13.73

B. Concentration, mg ^{246}Cm

At start			6.56	6.80
After irradiation			5.24	5.81
Date of measurement			Jan. 14, 1986	Mar. 27, 1990

C. Additional Postirradiation Data:

			Nov. 14, 1986	June 15, 1990
^{238}Pu, at. %			0.015	0.079
^{239}Pu, "			0.159	0.096
^{240}Pu, "			97.77	92.68
^{241}Pu, "			0.53	5.26
^{242}Pu, "			1.53	1.88
Pu wt., mg			Not measured	0.49

*Capsule lost during removal from fuel pin.

Sample data for ^{237}Np, Capsule Position 7

	At start	FP-1*	FP-2**	FP-4**
A. Isotopic Composition, at. %				
Date:	Aug 11, 1972			
236	--			
237	>99.99		>99.99	>99.99
238	--			
239	--			
B. Concentration, mg ^{237}Np				
At start			12.15	12.22
After irradiation			11.2	8.59
Date of measurement			Jan. 14, 1987	Jan. 14, 1992
C. Additional Postirradiation Data:				
			Nov. 21, 1986	Oct. 15, 1990
^{238}Pu, at. %			99.20	93.56
^{239}Pu, "			0.79	6.13
^{240}Pu, "			0.012	0.31
Pu wt., mg			0.48	3.30

* Capsule lost during removal from fuel pin.
** Assumed isotopic composition.

Sample data for ^{244}Cm, Capsule Position 8

A. Isotopic Composition, at. %

Date:	At start* June 16, 1981	FP-1** Aug. 23, 1984	FP-2 Nov. 19, 1986	FP-4***
243	0.016	--	--	
244	92.40	89.93	89.27	
245	1.162	2.78	2.95	
246	6.26	7.04	7.50	
247	0.101	0.182	0.198	
248	0.057	0.072	0.087****	

B. Concentration, mg ^{244}Cm

At start			8.26
After irradiation			6.08
Date of measurement			Jan. 14, 1987

C. Additional Postirradiation Data:

	June 16, 1981		Nov. 14, 1986
^{238}Pu, at. %	0.002		--
^{239}Pu, "	0.048		0.017
^{240}Pu, "	99.922		99.33
^{241}Pu, "	0.004		0.625
^{242}Pu, "	0.024		0.031
Pu wt., mg	0.73		2.07

* Starting ^{244}Cm contained 0.088 mg ^{240}Pu/mg of ^{244}Cm.

** Damaged during removal from fuel pin; no concentration measurement.

*** Lost during removal from fuel pin.

**** Incorrectly reported as 0.987 in Ref. 5, p. 42.

Sample data for ^{244}Cm, Capsule Position 9

A. Isotopic Composition, at. %

	At start*	FP-1**	FP-2	FP-4***
Date:	June 16, 1981	July 25, 1984	Nov. 20, 1986	July 13, 1990
243	0.016	--		-- --
244	92.40	90.04	89.28	78.75
245	1.162	2.74	2.94	11.77
246	6.26	6.96	7.51	8.61
247	0.101	0.19	0.20	0.75
248	0.057	0.07	0.074	0.12

B. Concentration, mg ^{244}Cm

At start		8.30	8.40	8.25
After irradiation		6.60	6.08	4.48
Date of measurement		Sept. 5, 1984	Jan. 14, 1987	March 26, 1990

C. Additional Postirradiation Data:

		Sept. 9, 1984	Nov. 20, 1986	June 18, 1990
^{238}Pu, at. %		<0.002	<0.002	0.058
^{239}Pu, "		0.018	0.017	0.021
^{240}Pu, "		99.04	99.31	93.83
^{241}Pu, "		0.91	0.63	5.75
^{242}Pu, "		0.031	0.038	0.34
Pu wt., mg		1.60	2.18	2.70 by RCA** 2.80 by IDMS**

* Starting ^{244}Cm contained 0.088 mg ^{240}Pu/mg of ^{244}Cm.

** Concentration results reported by two methods for quality assurance, radiochemical analysis and isotope dilution mass spectrometry.

Sample data for ^{243}Cm, Capsule Position 10

A. Isotopic Composition, at. %

	At start	FP-1	FP-2	FP-4
Date:	July 18, 1990	Aug. 9, 1984	Dec. 4, 1986	July 10, 1990
243	57.78	56.99	57.17	43.34
244	40.26	40.09	39.77	47.06
245	0.96	1.74	1.85	7.36
246	0.98	1.15	1.22	2.06
247	≤0.02	0.03	<0.05	0.16
248	≤0.01	<0.01	<0.05	0.02

B. Concentration, mg ^{243}Cm

At start		0.34	0.38	0.39
After irradiation		0.33	0.28	0.22
Date of measurement		July 24, 1984	Jan. 14, 1987	March 26, 1990

C. Additional Postirradiation Data:

None

Sample data for ^{238}U, Capsule Position 11

A. Isotopic Composition, at. %	At start	FP-1	FP-2	FP-4**
Date:	July 20, 1980	June 29, 1984	Sept. 18, 1986	July 20, 1990
233	<0.001	<0.001	0.001	<0.001
234	0.0015	0.0012	0.0016	0.002
235	0.044	0.047	0.042	0.031
236	<0.001	0.0006	0.0011	0.006
238	99.955	99.952*	99.955	99.961
B. Concentration, mg ^{238}U				
At start		9.86	10.33	9.81
After irradiation		9.64	9.37	1.75
Date of measurement		June 29, 1984	Sept. 18, 1986	
C. Additional Postirradiation Data:				
		June 29, 1984	Sept. 18, 1986	Aug. 13, 1990
^{238}Pu, at. %		0.012	<0.01	0.076
^{239}Pu, "		99.27	99.27	93.44
^{240}Pu, "		0.717	0.724	6.27
^{241}Pu, "		0.003	0.003	0.206
^{242}Pu, "		<0.001	<0.001	0.0058
Pu wt., mg		0.071	0.065	0.105

* Incorrectly reported as 99.940 in Ref. 5, p. 45.

** Portion of sample lost during dissolution.

FP-4 Sample data for ^{243}Am, Capsule Position 12

A. Isotopic Composition, at. %	At start	FP-1	FP-2	FP-4
Date:	Dec. 19, 1977	June 26, 1984	Dec. 2, 1986	May 21, 1990
241	0.012	<0.011	0.012	0.030*
242	<0.001	-	-	<0.001
243	99.988	99.988	99.987	99.970
B. Concentration, mg ^{243}Am				
At start		9.80	10.33	9.83
After irradiation		9.60	9.22	6.79
Date of measurement		Sept. 5, 1984	Aug. 14, 1987	March 27, 1990
C. Additional Postirradiation Data:				
		June 29, 1984	Sept. 18, 1986	June 11, 1990
^{238}Pu, at. %		0.45	0.12	0.13
^{239}Pu, "		20.68	12.87	2.02
^{240}Pu, "		78.87	87.01	95.15
^{241}Pu, "		**	**	2.22
^{242}Pu, "		**	**	0.20
^{244}Pu, "		**	**	0.28
Pu wt., mg		**	**	0.34
^{244}Cm, at. %		99.16	99.09	92.86
^{245}Cm, "		0.84	0.91	6.93
^{246}Cm, "		0.003	**	0.21
Cm wt., mg		0.34	0.31	2.46

* Suspect small ^{241}Pu isobaric contamination; no chemical separation made.

** Signal too weak for valid measurement.

FP-4 Sample data for ^{243}Am, Capsule Position 13

A. Isotopic Composition, at. %	At start	FP-1	FP-2	FP-4
Date:	Dec. 19, 1977	Aug. 23 1984	June 19, 1986	May 21, 1990
241	0.012	0.012	0.013	0.028*
242	<0.001	-	-	<0.001
243	99.988	99.988	99.987	99.972
B. Concentration, mg ^{243}Am				
At start		9.94	10.00	9.80
After irradiation		9.40	8.69	6.60
Date of measurement		Aug. 15, 1984	Aug. 14, 1987	March 27, 1990
C. Additional Postirradiation Data:				
		Aug. 23, 1984	June 19, 1987	June 20, 1990
^{238}Pu, at. %		**	0.44	0.13
^{239}Pu, "		**	12.58	2.06
^{240}Pu, "		**	86.98	95.23
^{241}Pu, "		**	**	2.17
^{242}Pu, "		**	**	0.13
^{244}Pu, "		**	**	0.28
Pu wt., mg		**	**	0.34
^{244}Cm, at. %		98.88	99.06	92.80
^{245}Cm, "		0.90	0.90	6.98
^{246}Cm, "		0.22	0.04	0.22
Cm wt., mg		0.31	0.28	2.56

* Suspect small ^{241}Pu isobaric contamination; no chemical separation made.

** Signal too weak for valid measurement.

FP-4 Sample data for ^{241}Am, Capsule Position 14

A. Isotopic Composition, at. %

Date:	At start Mar. 21, 1981	FP-1 June 29, 1984	FP-2 June 18, 1987	FP-4 June 1, 1990
241	99.995	99.33	99.34	94.26
242	<0.001	0.66	0.65	5.10
243	0.004	0.01	0.01	0.64

B. Concentration, mg ^{241}Am

	FP-1	FP-2	FP-4
At start	10.38	9.70	9.42
After irradiation	10.10	9.12	6.21
Date of measurement	Sept. 5, 1984	June 12, 1987	March 26, 1990

C. Additional Postirradiation Data:

	Sept. 5, 1984 *	Aug. 14, 1987 **	June 18, 1990 **
^{238}Pu, at. %	99.20	82.04	77.75
^{239}Pu, "	0.50	0.35	4.03
^{240}Pu, "	0.30	0.36	0.40
^{241}Pu, "	-	0.082	0.02
^{242}Pu, "	-	17.17	17.80
^{238}Pu wt., mg	0.27	0.28	1.92
^{244}Cm, at. %	-	-	95.28
^{245}Cm, "	-	-	4.16
^{246}Cm, "	-	-	0.48
^{247}Cm, "	-	-	0.01
^{248}Cm, "	-	-	0.07
^{242}Cm wt., mg	-	-	0.032

* FP-1: no Pu separation made from Am and Cm; unable to measure 241 and 242.

** FP-2, FP-4: Pu separated from Am and Cm; complete isotopic Pu made.

FP-4 Sample data for ^{241}Am, Capsule Position 15

A. Isotopic Composition, at. %	At start	FP-1	FP-2	FP-4
Date:	Mar. 21, 1981	Aug. 8, 1984	June 18, 1987	June 1, 1990
241	99.995	99.21	99.37	94.23
242	<0.001	0.78	0.62	5.12
243	0.004	0.01	0.01	0.65
B. Concentration, mg ^{241}Am				
At start		9.55	9.39	9.52
After irradiation		8.84	9.05	6.36
Date of measurement		Sept. 5, 1984	June 16, 1987	March 26, 1990
C. Additional Postirradiation Data:				
		Sept. 5, 1984*	Aug. 14, 1987**	June 20, 1990**
^{238}Pu, at. %		95.50	82.41	77.88
^{239}Pu, "		3.80	0.36	4.07
^{240}Pu, "		0.70	0.12	0.36
^{241}Pu, "		-	0.007	0.02
^{242}Pu, "		-	17.11	17.66
^{238}Pu wt., mg		0.25	0.28	1.92
^{244}Cm, at. %		-	-	95.72
^{245}Cm, "		-	-	4.08
^{246}Cm, "		-	-	1.03
^{247}Cm, "		-	-	0.03
^{248}Cm, "		-	-	0.14
^{242}Cm wt., mg		0.00004	0.00006	0.032

* FP-1: some contamination; also no Pu separation made from Am and Cm; unable to measure 241 and 242.

** FP-2, FP-4: Pu separated from Am and Cm; complete isotopic Pu made.

FP-4 Sample data for ^{242}Pu, Capsule Position 16

	At start	FP-1*	FP-2**	FP-4***
A. Isotopic Composition, at. %				
Date:	June 30, 1982	Sept. 17, 1984	Oct. 9, 1986	Aug. 14, 1990
238	0.124	0.124	0.019	0.09
239	0.383	3.256	0.83	0.25
240	1.023	1.990	0.98	0.90
241	0.093	0.126	0.083	0.11
242	97.950	94.041	97.48	98.30
244	0.427	0.462	0.514	0.35
B. Concentration, mg ^{242}Pu				
At start			2.09	2.05
After irradiation			1.67	0.78
Date of measurement			Oct. 30, 1986	Feb. 13, 1991
C. Additional Postirradiation Data:				
			Oct. 30, 1986	July 30, 1990
^{241}Am, at. %			8.40	0.91
^{243}Am, "			91.60	99.09
Am wt., mg			0.015	0.06

* Most of FP-1 sample was lost in dissolution: contamination likely, especially in 239, 240, and 241.
** some contamination of FP-2 suspected but of smaller magnitude than in FP-1.
*** Severe sample loss is suspected.

FP-4 Sample data for ^{244}Pu, Capsule Position 20

	At start	FP-1*	FP-2	FP-4
A. Isotopic Composition, at. %				
Date:	July 30, 1981		Oct. 16, 1986	Aug. 15, 1990
238	0.013		0.017	0.094
239	0.040		0.038	0.038
240	2.796		2.784	2.604
241	0.540		0.429	0.426
242	9.036		8.995	8.82
244	87.575		87.738	88.02
B. Concentration, mg ^{244}Pu				
At start			2.14	2.11
After irradiation			1.92	1.91
Date of measurement			Oct. 16, 1986	Feb. 13, 1991
C. Additional Postirradiation Data:				
				July 26, 1990
^{244}Cm, at. %				12.42
^{245}Cm, "				83.41
^{246}Cm, "				4.05
^{247}Cm, "				0.13
^{248}Cm, "				<0.01
Cm wt., mg				0.03

* Lost during removal from FP-1.

FP-4 Sample data for ^{240}Pu, Capsule Position 21

A. Isotopic Composition, at. %

	At start	FP-1	FP-2	FP-4
Date:	June 30, 1982	Sept. 17, 1984	Jan. 9, 1987	April 11, 1990
238	0.092	0.086	0.087	0.168
239	0.019	0.016	0.015	0.025
240	99.86	98.85	98.962	91.56
241	0.003	1.024	0.912	7.74
242	0.020	0.026	0.025	0.510
244	<0.001	<0.001	-0.003	<0.001

B. Concentration, mg ^{240}Pu

	At start	FP-1	FP-2	FP-4
At start		10.78	10.58	10.84
After irradiation		11.01	9.41	10.26
Date of measurement		Sept. 17, 1984	Jan. 9, 1987	Mar. 22, 1991

C. Additional Postirradiation Data:

	June 7, 1990
^{241}Am, at. %	97.48
^{242}Am, "	1.23
^{243}Am, "	1.29
Am wt., mg	0.135

FP-4 Sample data for ^{240}Pu, Capsule Position 22

	At start	FP-1	FP-2	FP-4
A. Isotopic Composition, at. %				
Date:	June 30, 1982	Sept. 17, 1984	Jan. 29, 1987	April 11, 1990
238	0.092	0.088	0.087	0.168
239	0.019	0.041	0.015	0.023
240	99.86	98.84	98.97	91.59
241	0.003	1.005	0.900	7.715
242	0.020	0.026	0.026	0.502
244	<0.001	<0.001	~0.001	<0.001
B. Concentration, mg ^{240}Pu				
At start		10.54	10.65	10.82
After irradiation		11.20	10.62	9.16
Date of measurement		Sept. 17, 1984	Jan. 29, 1987	Mar. 22, 1991
C. Additional Postirradiation Data:				
				June 8, 1990
^{241}Am, at. %				97.48
^{242}Am, "				1.23
^{243}Am, "				1.29
Am wt., mg				0.113

FP-4 Sample data for ^{239}Pu, Capsule Position 23

A. Isotopic Composition, at. %

	At start	FP-1*	FP-2	FP-4*
Date:	Aug. 11, 1972	Sept. 19, 1984	Feb. 20, 1987	April 17, 1990
238	<0.002	0.003	0.002	0.016
239	99.107	97.854	97.877	84.512
240	0.877	2.115	2.094	14.640
241	0.011	0.022	0.020	0.790
242	0.005	0.006	0.006	0.042
244	<0.005	<0.001	<0.001	<0.005

B. Concentration, mg ^{239}Pu

	FP-1*	FP-2	FP-4*
At start	7.99	7.92	8.47
After irradiation	*	7.32	3.55
Date of measurement	*	Feb. 20, 1987	April 17, 1990

C. Additional Postirradiation Data:

None

* Portion of FP-1 and FP-4 samples was lost in dissolution.

FP-4 Sample data for ^{241}Pu, Capsule Position 24

A. Isotopic Composition, at. %

Date:	At start April 23, 1982	FP-1 Sept. 19, 1984	FP-2 Mar. 11, 1987	FP-4 April 20, 1990
238	0.011	1.424	1.791	22.501
239	1.177	1.326	1.474	2.300
240	1.268	1.469	1.633	2.018
241	96.759	93.644	92.746	57.899
242	0.785	2.135	2.353	15.276
244	<0.005	<0.001	<0.001	0.006

B. Concentration, mg ^{241}Pu

At start	4.096	4.036	4.251
After irradiation	2.52	2.36	1.132
Date of measurement	Sept. 19, 1984	Mar. 11, 1987	Mar. 22, 1991

C. Additional Postirradiation Data:

	Aug. 6, 1984	Aug. 14, 1987	June 12, 1990
^{241}Am, at. %	99.43	99.52	94.30
^{242}Am, "	0.54	0.46	4.46
^{243}Am, "	0.03	0.02	1.24
^{241}Am wt., mg	1.60	1.87	1.75

FP-4 Sample data for ^{232}Th, Capsule Position 25

A. Isotopic Composition, at. %				
	At start	FP-1	FP-2	FP-4
Date:	June 30, 1981	Oct. 11, 1984	Mar. 16, 1987	Feb. 6, 1991
232	100.0	100.0	100.0	100.0
B. Concentration, mg ^{232}Th				
At start		17.77	17.88	18.80
After irradiation		17.70	17.70	18.11
Date of measurement		Oct. 11, 1984	Mar. 16, 1987	Feb. 6, 1991
C. Additional Postirradiation Data:				
		Oct. 11, 1984	Mar. 16, 1987	Oct. 19, 1990
^{233}U, at. %		99.45	99.60	95.25
^{234}U, "		0.55	0.40	4.54
^{235}U, "		-	-	0.21
^{236}U, "		-	-	<0.01
^{238}U, "		-	-	<0.01
^{233}U wt., mg		0.15	0.16	1.40

FP-4 Sample data for ^{236}U, Capsule Position 26

A. Isotopic Composition, at. %

	At start	FP-1	FP-2	FP-4*
Date:	July 20, 1981	June 29, 1984	Sept. 18, 1986	Aug. 16, 1990
233	0.207	0.213	0.152	0.09
234	0.122	0.123	0.123	0.22
235	9.352	9.00	9.04	6.21
236	88.929	89.20	89.25	91.51
238	1.390	1.46	1.43	1.97

B. Concentration, mg ^{236}U

At start		7.91	7.92	7.96
After irradiation		8.26	7.41	3.69
Date of measurement		June 29, 1984	Nov. 10, 1986	Feb. 2, 1991

C. Additional Postirradiation Data:

		June 29, 1984	Sept. 23, 1986	Aug. 15, 1990
^{238}Pu, at. %		69.50	66.10	90.65
^{239}Pu, "		30.20	33.50	8.85
^{240}Pu, "		0.30	0.40	0.479
^{241}Pu, "		-	-	0.014
^{242}Pu, "		-	-	0.003
Pu wt., mg		0.003	0.002	0.09
^{237}Np wt., mg		0.09	0.12	0.68

* Severe sample loss is suspected.

FP-4 Sample Data for ^{234}U, Capsule Position 27

A. Isotopic Analysis, at. %

Date:	At start Mar. 5, 1982	FP-1 June 29, 1984	FP-2 Sept. 19, 1986	FP-4 Nov. 26, 1990
233	0.0022	0.052	0.036	0.08
234	99.766	98.37	98.45	88.19
235	0.168	1.46	1.43	10.91
236	0.040	0.084	0.048	0.85
238	0.024	0.037	0.04	0.036

B. Concentration, mg ^{234}U

At start		3.44	3.27	3.50
After irradiation		3.93	3.23	3.33
Date of measurement		June 29, 1984	Sept. 19, 1986	Nov. 26, 1990

C. Additional Postirradiation Data:

	June 29, 1984	Sept. 19, 1986	Nov. 26, 1990
^{238}Pu, at. %	92.0	92.60	90.70
^{239}Pu, "	7.6	6.70	8.73
^{240}Pu, "	0.4	0.65	0.54
^{241}Pu, "	--	0.04	0.022

FP-4 Sample Data for ^{235}U, Capsule Position 28

A. Isotopic Analysis, at. %

	At start	FP-1	FP-2	FP-4
Date:	Aug. 11, 1972	June 29, 1984	Aug. 22, 1986	Nov. 29. 1990
233	<0.001	<0.01	<0.01	<0.01
234	0.034	0.036	0.039	0.078
235	99.89	98.69	98.66	85.51
236	0.025	1.18	1.17	14.34
238	0.053	0.099	0.121	0.066

B. Concentration, mg ^{235}U

At start		8.53	8.11	8.37
After irradiation		8.04	7.36	4.96
Date of measurement		June 29, 1984	Aug. 22, 1986	Jan. 25, 1991

C. Additional Postirradiation Data:

None

Sample Data for ^{231}Pa, Capsule Position 29

A. Isotopic Composition, at. %

No measurement required: ^{231}Pa, 100%.

B. Concentration, mg ^{231}Pa

	FP-1	FP-2*	FP-4***
At start	2.89		2.512
After irradiation	3.80		0.90
Date of measurement	Sept. 5, 1984		Jan. 14, 1992

C. Additional Postirradition Data:

	Sept. 5, 1984		May 31, 1990
^{232}U** wt., mg	0.10		0.41

* Sample lost in dissolving.
** Incorrectly reported as ^{233}U in Ref. 5, p. 60.
*** Some sample loss is suspected.

Sample Data for ^{238}Pu, Capsule Position 30

A. Isotopic Composition, at. %

	At start	FP-1*	FP-2**	FP-4***
Date:	June 20, 1972		May 12, 1987	April 16, 1990
238	99.39		98.07	86.74
239	0.279		1.54	11.89
240	0.319		0.367	1.282
241	<0.01		0.006	0.068
242	0.018		0.019	0.026
244	<0.001		<0.001	<0.001

B. Concentration, mg ^{238}Pu

	FP-1*	FP-2**	FP-4***
At start	2.91	3.27	3.11
After irradiation	*	**	0.65****
Date of measurement	*	Mar. 12, 1987	Mar. 26, 1990

C. Additional Postirradition Data:

None

* Not analyzed; damaged while removing from FP-1.
** Portion lost in dissolution.
*** Severe sample loss is suspected.
**** From alpha spectrometry. Isotope dilution mass spectrometry gave 0.80 mg.

Sample Data for ^{230}Th, Capsule Position 31

A. Isotopic Composition, at. %

	At start	FP-1*	FP-2**	FP-4
Date:	Mar. 5, 1982		Jan. 20, 1987	Feb. 20, 1991
230	89.47		89.37	88.92
232	10.53		10.63	11.08

B. Concentration, mg ^{230}Th

	FP-1*	FP-2**	FP-4
At start	2.92	2.93	2.88
After irradiation	*	2.66	2.54
Date of measurement	*	Jan. 20, 1987	Feb. 20, 1991

C. Additional Postirradition Data:

None

* Lost in removal from FP-1.

** Small loss of sample from bumping during dissolution.

FP-4 Sample Data for ^{233}U, Capsule Position 32

A. Isotopic Composition, at. %

	At start	FP-1	FP-2*	FP-4*
Date:	Mar. 5, 1982	Nov. 10, 1984	Sept. 3, 1986	Dec. 7, 1990
233	99.887	99.41	99.36	92.01
234	0.052	0.55	0.58	7.34
235	0.0032	0.005	0.009	0.54
236	0.0018	0.001	0.002	0.031
238	0.056	0.031	0.051	0.080

B. Concentration, mg ^{233}U

At start		7.92	7.78	7.65
After irradiation		7.45	*	*
Date of measurement		Nov. 10, 1984		

C. Additional Postirradition Data:

* Portion lost in dissolution; isotopic data excellent

Appendix C

ANALYTICAL DATA FOR IRRADIATED DOSIMETERS

Sample Data for ^{235}U, Capsule Position 1

A. Isotopic Composition, at. %

Date:	At Start Aug. 11, 1972	FP-1 Nov. 27, 1984	FP-2 Oct. 7, 1986	FP-4 Aug. 8, 1991
234	0.034	0.039	0.040	0.072
235	99.89	98.35	98.38	87.42
236	0.025	1.55	1.52	12.51
238	0.053	0.066	0.060	

B. Concentration, mg ^{235}u

At start		1.09	1.14	1.18
After irradiation		1.09	1.09	0.40
Date of measurement		Nov. 27, 1984	Oct. 7, 1986	Oct. 2, 1991

C. Additional Postirradition Data:

None

Note: FP-4 dosimeters (^{235}U and ^{238}U) in Capsule 1 could not be physically separated before dissolving; therefore, isotopic results assume that all isotopes other than ^{238}U are derived from the ^{235}U dosimeter.

Sample Data for ^{238}U, Capsule Position 1

A. Isotopic Composition, at. %

	At start	FP-1	FP-2	FP-4
Date:	Aug. 11, 1972	Nov. 26, 1984	Oct. 7, 1986	Aug. 8, 1991
234	<0.0001	<0.0005	<0.0005	-
235	0.0012	0.0040	0.0012	-
236	<0.0001	<0.0005	<0.0005	-
238	99.999	99.996	99.999	~99.99

B. Concentration, mg ^{238}u

	FP-1	FP-2	FP-4
At start	1.01	1.15	1.18
After irradiation	1.07	1.16	0.60
Date of measurement	Nov. 26, 1984	Oct. 7, 1986	Oct. 24, 1991

C. Additional Postirradition Data:

	Nov. 26, 1984	Oct. 7, 1986	Aug. 8, 1991
^{238}Pu, at. %	-	-	1.072
^{239}Pu, "	99.20	99.21	93.14
^{240}Pu, "	0.80	0.79	5.64
^{241}Pu, "	0.004	0.003	0.140
^{242}Pu, "	0.001	0.001	0.006
Pu, wt., mg	0.007	0.009	0.033

Note: FP-4 dosimeters (^{235}U and ^{238}U) in Capsule 1 could not be physically separated before dissolving; therefore, results for ^{238}U assume (1) only ^{238}U present in terms of U, and (2) all Pu comes from ^{238}U irradiation.

Sample Data for ^{239}Pu, Capsule Position 2

A. Isotopic Composition, at. %

	At start	FP-1	FP-2*	FP-4
Date:	Aug. 11, 1972	Dec. 17, 1984		Aug. 23, 1991
238	<0.0002	0.002		21.43
239	99.107	97.898		68.26
240	0.877	2.072		9.88
241	0.011	0.021		0.40
242	0.005	0.061		0.026

B. Concentration, mg ^{239}Pu

	FP-1	FP-4
At start	1.24	1.20
After irradiation	0.58	0.77
Date of measurement	Jan. 3, 1985	Aug. 1, 1991

C. Additional Postirradition Data:

None

Note: FP-4 dosimenters (^{237}Np and ^{239}Pu) in capsule 2 could not be physically separated before dissolving; therefore, results show the combined ^{239}Pu left from the original dosimeter and an additional amount produced from the neutron bombardment of the original ^{237}Np.

* Lost in handling.

Sample Data for ^{237}Np, Capsule Position 2

A. Isotopic Composition, at. %

Date:	At start Aug. 21, 1972	FP-1*	FP-2*	FP-4*
237	>99.99	>99.99	>99.99	>99.99

B. Concentration, mg ^{237}Np

	FP-1*	FP-2*	FP-4*
At start	1.14	1.07	1.04
After irradiation	0.96	0.95	0.62
Date of measurement	Jan. 21, 1985	Aug. 29, 1988	Jan. 10, 1992

C. Additional Postirradition Data:

	Nov. 26, 1984	Aug. 29, 1988	Aug. 23, 1991
^{238}Pu wt., mg	0.045	0.044	0.24

Note: Although FP-4 dosimeters (^{237}Np and ^{239}Pu) in capsule 2 could not be physically separated before dissolving, the ^{238}Pu should be from the ^{237}Np dosimeter only.

*Assumed isotopic compositions.

Sample Data for ^{235}U, Capsule Position 17

A. Isotopic Composition, at. %

Date:	At start Aug. 11, 1972	FP-1 Nov. 27, 1984	FP-2 Aug. 9, 1988	FP-4 Aug. 23, 1991
234	0.034	0.038	0.040	0.067
235	99.89	98.57	98.54	85.46
236	0.025	1.325	1.31	14.47
238	0.053	0.070	0.11	-

B. Concentration, mg ^{235}U

At start		1.05	1.18	1.11
After irradiation		1.03	1.14	0.34
Date of measurement		Nov. 27, 1984	Aug. 9, 1988	Oct. 29, 1991

C. Additional Postirradition Data:

None

Note: FP-4 dosimeters (^{235}U and ^{238}U) in capsule 17 could not be physically separated before dissolving; therefore, isotopic results assume that all isotopes other than ^{238}U are derived from the ^{235}U dosimeter.

Sample Data for ^{238}U, Capsule Position 17

A. Isotopic Composition, at. %

	At start	FP-1	FP-2*	FP-4
Date:	Aug. 11, 1972	Nov. 29, 1984		Aug. 23, 1991
234	<0.0001	<0.0005		-
235	0.0012	0.0018		-
236	<0.0001	<0.0005		-
238	99.999	99.998		~99.99

B. Concentration, mg ^{238}U

	At start	FP-1	FP-2*	FP-4
At start		1.13		0.98
After irradiation		0.99		0.50
Date of measurement		Nov. 29, 1984		Oct. 29, 1991

C. Additional Postirradition Data:

	At start	FP-1	FP-2*	FP-4
		Nov. 29, 1984		Oct. 29, 1991
^{238}Pu, at. %		-		1.572
^{239}Pu, "		99.31		91.79
^{240}Pu, "		0.69		6.45
^{241}Pu, "		0.004		0.180
^{242}Pu, "		0.001		0.013
Pu wt., mg		0.006		0.031

Note: FP-4 dosimeters (^{235}U and ^{238}U) in capsule 17 could not be physically separated before dissolving; therefore, results for ^{238}U assume (1) only ^{238}U present in terms of U, and (2) all Pu starts from ^{238}U irradiation.

* Lost in handling.

Sample Data for ^{239}Pu, Capsule Position 18

A. Isotopic Composition, at. %

Date:	At start Aug. 11, 1972	FP-1 Dec. 17, 1984	FP-2*	FP-4 Oct. 29, 1991
238	<0.002	-		25.72
239	99.107	97.81		63.14
240	0.877	2.16		10.57
241	0.011	0.022		0.54
242	0.005	0.006		0.034

B. Concentration, mg ^{239}Pu

	At start	FP-1	FP-2*	FP-4
At start		1.19		1.12
After irradiation		0.48		0.68
Date of measurement		Dec. 17, 1984		Oct. 29, 1991

C. Additional Postirradition Data:

None

Note: FP-4 dosimeters (^{237}Np and ^{239}Pu) in capsule 18 could not be physically separated before dissolving; therefore, results show the combined ^{239}Pu left from the original dosimeter and an additional amount produced from the neutron bombardment of the original ^{237}Np.

* Lost in handling.

Sample Data for ^{237}Np, Capsule Position 18

	At start	FP-1*	FP-2**	FP-4*
A. Isotopic Composition, at. %				
Date:	Aug. 21, 1972			
	>99.99	>99.99		>99.99
B. Concentration, mg ^{237}Np				
At start		1.12		1.12
After irradiation		1.00		0.62
Date of measurements		Feb. 8, 1985		Jan. 10, 1992
C. Additional Postirradition Data:				
		Dec. 14, 1984		Aug. 23, 1991
^{238}Pu wt., mg		0.039		0.27

Note: Although FP-4 dosimeters (^{237}Np and ^{239}Pu) in capsule 18 could not be physically separated before dissolving, the ^{238}Pu should be from the ^{237}Np dosimeter only.

* Assumed isotopic composition.
** Lost in handling.

Sample Data for ^{235}U, Capsule Position 33

A. Isotopic Composition, at. %

	At start	FP-1	FP-2	FP-4*
Date:	Aug. 11, 1972	Dec. 11, 1984	Aug. 11, 1988	
234	0.034	0.037	0.040	
235	99.89	98.73	98.73	
236	0.025	1.15	1.13	
238	0.053	0.085	0.100	

B. Concentration, mg ^{235}U

	At start	FP-1	FP-2	FP-4*
At start		1.07	1.16	
After irradiation		1.06	1.12	
Date of measurement		Dec. 11, 1984	Aug. 11, 1988	

C. Additional Postirradition Data:

None

---

* Lost in handling.

Sample Data for ^{238}U, Capsule Position 33

A. Isotopic Composition, at. %				
Date:	At start Aug 11, 1972	FP-1 Dec. 7. 1984	FP-2 Aug. 11, 1988	FP-4*
234	<0.0001	<0.0005	<0.0005	
235	0.0012	0.0026	0.0020	
236	<0.0001	0.0005	<0.0005	
238	99.999	99.997	99.998	
B. Concentration, mg ^{238}U				
At start		1.01	0.97	
After irradiation		1.13	0.96	
Date of measurement		Dec. 7, 1984	Aug. 11, 1988	
C. Additional Postirradition Data:				
^{239}Pu, at. %		99.38	99.38	
^{240}Pu, "		0.62	0.62	
^{241}Pu, "		0.003	0.002	
^{242}Pu, "		0.001	≤0.001	
Pu, wt., mg		0.006	0.006	

* Lost in handling.

Sample Data for ^{239}Pu, Capsule Position 34

A. Isotopic Composition, at. %

	At start	FP-1	FP-2*	FP-4
Date:	Aug. 11, 1972	Dec. 17, 1984		Oct. 7, 1991
238	<0.002	-		25.20
239	99.107	97.58		63.83
240	0.877	2.38		10.40
241	0.011	0.027		0.53
242	0.005	0.006		0.032

B. Concentration, mg ^{239}Pu

	At start	FP-1	FP-2*	FP-4
At start		1.20		1.18
After irradiation		0.79		0.69
Date of measurement		Dec. 17, 1984		Oct. 29, 1991

C. Additional Postirradition Data:

None

Note: FP-4 dosimeters (^{237}Np and ^{239}Pu) in capsule 34 could not be physically separated before dissolving; therefore, results show the combined ^{239}Pu left from the original dosimeter and an additional amount produced from the neutron bombardment of the original ^{237}Np.

* Lost in handling.

Sample Data for ^{237}Np, Capsule Position 34

	At start	FP-1*	FP-2**	FP-4*
A. Isotopic Composition, at. %				
Date:	Aug. 21, 1972			
237	>99.99	>99.99		>99.99
B. Concentration, mg ^{237}Np				
At start		1.13		1.18
After irradiation		1.00		0.59
Date of measurement		Jan. 21, 1985		Jan. 10, 1992
C. Additional Postirradition Data:		Dec. 14, 1984		Aug. 23, 1991
^{238}Pu wt., mg		0.040		0.26

Note: Although FP-4 dosimeters (^{237}Np and ^{239}Pu) in capsule 34 could not be physically separated before dissolving, the ^{238}Pu should be from the ^{237}Np dosimeter only.

* Assumed isotopic composition.
** Lost in handling.

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