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FINAL REPORT FOR THE LIGHT WATER BREEDER REACTOR
PROOF-OF-BREEDING ANALYTICAL SUPPORT PROJECT

by

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ABSTRACT

The technology of breeding ^{233}U from ^{232}Th in a light water reactor is being developed and evaluated by the Westinghouse Bettis Atomic Power Laboratory (BAPL) through operation and examination of the Shippingport Light Water Breeder Reactor (LWBR). Bettis is determining the end-of-life (EOL) inventory of fissile uranium in the LWBR core by nondestructive assay of a statistical sample comprising approximately 500 EOL fuel rods. This determination is being made with an irradiated-fuel assay gauge based on neutron interrogation and detection of delayed neutrons from each rod. The EOL fissile inventory will be compared with the beginning-of-life fissile loading of the LWBR to determine the extent of breeding.

In support of the BAPL proof-of-breeding (POB) effort, Argonne National Laboratory (ANL) carried out destructive physical, chemical, and radiometric analyses on 17 EOL LWBR fuel rods that were previously assayed with the nondestructive gauge. The ANL work included measurements on the intact rods; shearing of the rods into pre-designated contiguous segments; separate dissolution of each of the more than 150 segments; and analysis of the dissolver solutions to determine each segment's uranium content, uranium isotopic composition, and loading of selected fission products. This report describes the facilities in which this work was carried out, details operations involved in processing each rod, and presents a comprehensive discussion of uncertainties associated with each result of the ANL measurements.

Most operations were carried out remotely in shielded cells. Automated equipment and procedures, controlled by a computer system, provided error-free data acquisition and processing, as well as full replication of operations with each rod. Despite difficulties that arose during processing of a few rod segments, the ANL destructive-assay results satisfied the demanding needs of the parent LWBR-POB program.

SUMMARY

As part of a national program to develop the technology for breeding ^{233}U from ^{232}Th in a light water reactor, Argonne National Laboratory (ANL) has destructively analyzed 17 fuel rods from the end-of-life (EOL) core of the Light Water Breeder Reactor (LWBR), which operated at Shippingport, PA, from September 1977 through October 1982. These rods were part of a larger sample (~500 rods) that was to be nondestructively assayed by the Bettis Atomic Power Laboratory (BAPL) by means of its specially designed delayed-neutron device, designated the Production Irradiated Fuel Assay Gauge (PIFAG); BAPL is operated by the Westinghouse Electric Corporation for the Department of Energy Division of Naval Reactors (DOE-NR). The PIFAG assay determined the fissile uranium loading of each rod in the sample. Using statistical methods, BAPL intends to estimate the fissile inventory of the entire LWBR core (more than 17,000 rods). The results of the ANL analyses, which were performed on rods previously assayed with the PIFAG, were used by BAPL to obtain correction and calibration factors related to the PIFAG measurements and to corroborate the nondestructively determined rod fissile loadings. Breeding performance of the LWBR will be measured by BAPL through a comparison of the EOL fissile inventory with the known, beginning-of-life (BOL), fissile loading of the reactor.

The LWBR fuel rods were processed at ANL over a period of about 16 months, beginning in August 1984. The bulk of the work was done remotely, in shielded cells, using equipment and procedures that were developed and qualified specifically for this purpose prior to the onset of work with the EOL fuel rods. A computer system was used for process control, as well as for data acquisition, reduction, and management. Processing the rods included (1) making physical measurements to determine rod weight, length, and surface temperature; (2) shearing each rod into precisely characterized segments; (3) dissolving each of the 152 segments (135 fuel-bearing and 17 plenum segments); and (4) analyzing solutions for uranium isotopes and selected fission products. Fission gas (Xe and Kr) present in each rod's void volume was determined, as was the quantity of fission gas released during shearing and dissolution of each individual segment. The bulk analytical residues (mainly dissolver solution) were disposed of by conversion to a cement waste form and transportation to an approved site away from ANL. This final step served as a demonstration for the preparation of a waste package acceptable to the federal repository [Waste Isolation Pilot Plant (WIPP)] for transuranic (TRU) waste.

Because of the small magnitude of the breeding gain (~1.5%) expected from the LWBR and the relatively small number of fuel rods processed at ANL, exacting requirements were established by BAPL for the precision and accuracy of most of the measurements and analyses performed in the destructive assay. Through careful planning of the EOL operations and design of equipment and procedures (a listing of patents issued for inventions related to this work is given at the end of the report), together with extensive development and characterization of the operating systems once they were in place, we entered the EOL campaign with a high level of confidence that these requirements would be satisfied. Operations involved in processing the LWBR fuel rods and performance of the ANL systems during the processing are briefly described below for several key areas. Quoted figures of merit for the individual measurement results were derived from a comprehensive analysis of errors in which random and systematic errors associated with specific operations were evaluated from data obtained during processing of the EOL fuel rods or from pre-EOL characterization and

qualification tests performed with the EOL systems. When available, EOL data were always given preference in these evaluations. Uncertainties in individual results were estimated by conventional error-propagation techniques.

As the following discussion shows, execution of the EOL campaign proved not to be free of problems, but the problems that arose were remarkably few considering the difficulties associated with analytical operations performed in a hot-cell environment and the scheduler pressures of shearing 17 irradiated fuel rods into segments, carrying out 195 dissolutions (including rod segments and blanks), and performing several thousand analyses on the dissolver solutions and gas samples in a 16-month period. In general, project error requirements were met. In a few instances, unpredictable behavior of the irradiated rod materials or operational mishaps caused larger-than-normal errors or uncertainties to be associated with particular measured values. Also, a few of the measurement systems did not perform quite as well as expected from our pre-EOL characterization work. However, even in those few cases where specific limits to allowable error were exceeded, either additional data were available to permit BAPL's accounting for the errors in their final evaluation of the data or the uncertainties associated with the deviant results could be shown to be small enough that the needs of the parent program would be met.

Physical Measurements on Unsheared Rods. Physical measurements that were made for each fuel rod included weighing the rod, measuring its length, and determining its surface temperature. Fuel rods were weighed to the nearest 0.1 g, both before and after the rod's plenum was punctured to permit collection, measurement, and sampling of the plenum gas. Bias in each fuel rod weighing was rendered negligible relative to the resolution of the rod-weighing balance by calibrating the balance with a National Bureau of Standards (NBS)-traceable standard weight and was further demonstrated to be less than the resolution of the balance by weighing a transfer standard that was carefully compared to NBS-calibrated weights outside the hot cell. The limit to allowable bias in the fuel rod weighing was 0.1 g, and this requirement was met.

The standard deviation of an individual rod weighing was estimated from duplicate (before and after puncture) weighings and had the value 0.13 g. This value exceeds the corresponding limit to allowable error (0.10 g) by only a slight amount and is believed to satisfy the intent if not the letter of the requirement. If the fuel rod weight were computed as the average of the before- and after-puncture weights, its standard deviation would be reduced by the factor $\sqrt{2}/2$, to 0.09 g, and the requirement would be met.

Rod length was determined from measurements with the feed/measurement system on a full-scale shear (FSS), which is described below. Potential bias in the (~120-in.) rod length values obtained in this way arose from uncertainties in the length of a standard gauge rod used in calibrating the system and from such sources as nonlinearity of the ball screw that drives the FSS feed system. The limit to allowable bias in the rod-length determination was 0.001 in. On the basis of data obtained during pre-EOL characterization tests, which utilized a laser-based displacement monitoring system for assessing performance of the feed system, we estimated the limit to probable bias in the rod-length determination at 0.0004 in., a value that easily satisfies the corresponding requirement. The standard deviation of the rod length measurement was similarly estimated as 0.0013 in., well within the limit (0.005 in.) allowed for this parameter.

The surface temperature of each fuel rod was measured to the nearest 0.1°C by means of resistance-type (RTD) sensors attached to positioners that held the rod in position on the shear bed. The temperature was measured and reported in two ways. In one case, an average of the readings from three different sensors at different locations on the rod was taken while the rod length was being measured. In the other case, a single sensor in contact with the rod was automatically read each time the rod was indexed forward into position for shearing. The sensor readings (generally several hundred separate values) provided a profile of rod surface temperature along the entire rod length. A plot of surface temperature vs. location on the rod was generated for each fuel rod by computer. No requirements were established in regard to the surface-temperature measurements; however, of interest was the excellent qualitative relationship that we noted between each rod's surface-temperature profile and its axial profile of fission-product content determined by analysis.

Shearing of the Fuel Rods. Shearing operations performed on each fuel rod were designed to accomplish three objectives: (1) to divide the rod into contiguous segments whose boundaries were located precisely at positions specified in detailed cutting plans provided by BAPL, (2) to comminute the fuel material in each segment for efficient subsequent dissolution, and (3) to collect each sheared segment with high recovery. Four sizes of rods were sheared: reflector, standard blanket, power-flattening blanket (PFB), and seed. The diameters of these rods were 0.832 in., 0.572 in., 0.527 in., and 0.306 in., respectively.

The actual shearing of each fuel rod was done under full computer control by means of an automated hydraulic FSS having a feed/measurement system capable of positioning the rod for cutting to within 0.001 in. of a desired location. Segments were sheared from the rod as a series of short cuts, each having a length-to-diameter ratio of two, to enhance pulverization of the ceramic fuel and separation of the fuel and cladding. Sheared material from each segment was collected directly in a preweighed aluminum can. When a designated segment boundary was approached, the length of the final cut was recalculated to account for rod slippage and thermal expansion of the shear feed system. The final cut was then made, the as-cut location of the segment boundary was determined, and the sample collection can was removed and capped. The capped aluminum cans were weighed, in-cell, to the nearest 0.001 g (0.01 g if the segment weight exceeded 286 g). Material balance calculations, which compared the sum of the segment weights for a given rod with the weight of the unsheared rod, measured the weight of material (primarily pulverized fuel) lost during shearing.

The project requirements established limits to allowable errors in the location of segment boundaries relative to the locations specified by BAPL, the determination of segment lengths, and the measurement of segment weights. For the boundary locations and segment length measurements, separate requirements were specified for the cladding and fuel components of a given segment to account for the different shearing characteristics of these two materials.

Under normal conditions, the FSS was capable of easily meeting the requirements on segment boundary location, which stipulated that the segment boundary in the cladding be located within 0.010 in. of the position specified by the shearing plan. Behavior of the LBWR fuel rods during shearing was, however, not ideal. Most of these rods showed some degree of slippage in the

carriage clamp jaws, as the rod was indexed forward for cutting, and occasional movement (kickback) of the spring-loaded jaws during the shear stroke. In most instances, these movements were compensated for in the computerized shearing procedure and did not affect the results of the shearing operations. When, however, kickback of the rod occurred during a segment boundary cut, an error in the segment boundary location resulted. Such was the case for eight (approximately 6%) of the 135 boundary cuts that were made in shearing the fuel-bearing portions of the LWBR fuel rods. Seven of the eight discrepant boundary locations occurred for cuts associated with one or the other of two reflector-type rods processed in the campaign. Kickback of these larger-diameter rods (0.832 in. as compared to 0.572 in. for the next smallest rod type) was exceptionally frequent and severe, causing displacements as large as 0.034 in. between the desired and as-cut segment boundaries.

Because the boundary location errors that occurred during the EOL campaign were few and resulted from the unpredictable shearing behavior of the irradiated Zircaloy cladding, BAPL indicated early in the campaign that performance of the FSS in locating the segment boundaries was acceptable on a best-effort basis. Moreover, because the actual locations of the segment boundaries were reliably known from our measurements (in the cladding: bias less than 0.004 in., standard deviation of 0.0013 in.; in the fuel: bias less than 0.0028 in., standard deviation of 0.0050 in.), deviations between the requested and as-cut locations could be accounted for in BAPL's final evaluation of the shearing data. Thus, although not on a par with what was expected on the basis of pre-EOL tests with nonirradiated, Zircaloy-clad, dummy rods, performance of the FSS in locating segment boundaries with the LWBR fuel rods was satisfactory overall.

The length of each individual segment was computed as the difference between the locations of the as-cut boundaries that defined the two ends of the segment. When this difference is calculated, bias errors in the measured boundary locations cancel because they are virtually independent of the segment boundary locations and the length of the segment. Thus, residual bias in the calculated segment lengths may be considered inconsequential. Limits to allowable bias were 0.001 in. for cladding segment length and 0.005 in. for fuel segment length. These requirements were easily met. Standard deviations of the segment length determinations were estimated to be 0.0018 in. for cladding segments and 0.0071 in. for fuel segments. These values demonstrate that the limiting standard deviations (0.005 in. for cladding, 0.015 in. for fuel) allowed by project requirements were not exceeded; hence, this requirement was also met.

Weights of the individual rod segments were calculated as the difference between the measured weight of a particular aluminum can when it contained the segment and when it was empty. All of the empty cans were weighed on the same 300-g capacity balance to the nearest 0.001 g. Full cans weighing 300 g or less (segment weight ≤ 286 g) were also weighed on this balance, while those weighing more than 300 g were weighed on a 2000-g capacity balance to the nearest 0.01 g. Calibration of each balance with NBS-traceable standard weights and verification of their calibration by means of transfer standards ensured that bias in the weighings was negligible relative to the balance resolution, and that bias requirements (0.001 g for segments ≤ 286 g; 0.01 g for segments > 286 g) were satisfied.

Standard deviations of the segment weights were estimated from replicate weighing data on the empty and full cans. For segments weighing ≤ 286 g (allowed standard deviation 0.005 g), the resulting standard deviation estimate had the value 0.0036 g and was well within the allowed limit. For segments weighing more than 286 g (allowable standard deviation also 0.005 g), the corresponding uncertainty was 0.0063 g, slightly larger than the limit allowed by project requirements. We knew at the outset of the EOL campaign that this particular requirement, which calls for a standard deviation equal to only one-half the resolution of the balance used for the weighings, would be difficult to meet. Because pre-EOL characterization testing of the weighing systems indicated that meeting the requirement was probable, however, ANL did not request its re-evaluation by BAPL. In spite of not meeting requirements, we believe that this observed standard deviation for the determination of segment weights greater than 286 g is the best that could be achieved with equipment that was commercially available when the ANL facilities were set up and, therefore, that it conforms to the intent of the requirement.

High recovery of the sheared fuel belonging to a given fuel rod is important because it reduces potential errors associated with fuel-loss corrections that BAPL applied to the rod assay results. If the magnitude of these corrections is kept small, uncertainties in the corrections are also small. Because the effects of rod irradiation on the shearing behavior of fuel material (and, hence, on fuel loss) were recognized as unpredictable, acceptance criteria for fuel recovery during shearing were specified in the project requirements in terms of minimum acceptable performance levels based on studies involving nonirradiated materials. These specifications called for an overall rod recovery fraction of the fuel material not less than 99.75%. Pre-EOL studies with nonirradiated test rods showed that, in fact, fuel recovery during shearing of thorium fuel pellets consistently exceeded 99.9%. In effect, fuel recovery in shearing the irradiated LWBR fuel rods was on a best-effort basis. However, we anticipated that the level of performance would be at least consistent with the 99.75% minimum criterion applied to the nonirradiated test rods.

Calculation of the actual fuel recovery fraction for a given rod requires knowledge of the total weight of fuel that the rod contained initially and was left for BAPL to carry out. However, using approximate oxide fuel weights that BAPL provided on the shearing diagram for each LWBR rod, we were able to compute approximate values for this parameter. Based on these approximate calculations, fuel recovery for 16 of the 17 LWBR fuel rods was indeed consistent with the target recovery, having values ranging from 99.73 to 99.97%. The lower recovery (99.36%) obtained for the remaining rod was linked to the use of smaller (3-in. vs. 6-in. tall) aluminum cans in collecting the sheared segments. The smaller cans were used for the one rod only.

Segment Dissolution. Each sheared segment, including the aluminum can used to collect the segment, was individually dissolved in one or the other of two virtually independent dissolver systems, which were installed in one of the project hot cells. The heart of each dissolver system was a tantalum primary vessel in a stainless steel secondary vessel for high-temperature, high-pressure dissolution of the refractory fuel material in the segments. In general, low-uranium-bearing segments were dissolved in one dissolver system, and segments having higher uranium loadings were dissolved in the second system. The overall dissolution sequence was carried out under full computer control on a round-the-clock basis.

It included a 4-h primary dissolution in Thorex solution (13.6M HNO_3 , 0.06M HF) followed by a dilute nitric acid rinse, then a 3-h secondary dissolution in Thorex solution containing 0.06M Al^{3+} (Thorex-Al) followed by a reflux rinse and a cold rinse with dilute nitric acid. Operating conditions for both dissolution periods were 195°C and 120 psig. The batches of primary and secondary dissolver solution and the several rinses were combined in a blend tank and thoroughly mixed prior to sampling. Gases released during the dissolution were retained in an off-gas holdup system and subsequently analyzed to determine their fission gas content.

Three sets of solution samples were taken for each segment. The first set (two samples) was taken from the secondary dissolver solution as it was being transferred to the blend tank. Measurement of the uranium in these samples allowed assessment of the completeness of fuel dissolution in the primary contacting of the segments with Thorex solution. The other two sample sets (four samples each) were withdrawn from the blend tank, one before ("unspiked" samples) and one after ("spiked" samples) a spike containing a known weight of NBS Standard Sample 950a (primarily ^{238}U) was added to the solution. The spiking procedure was part of the analysis to determine total uranium in the segment by the Mass Spectrometric Isotope Dilution (MSID) method. Half of the samples in each set were analyzed, while the other half were placed in archive storage.

The purpose of the segment dissolution was to convert fuel material contained in the segment into a form where it could be fully homogenized and conveniently analyzed. To fulfill this purpose, each segment had to be quantitatively dissolved and the fuel-bearing solution had to be made available in its entirety for analysis. In addition, carryover of dissolved fuel from one segment into the dissolution of a subsequent segment dissolved in the same system had to be at insignificant levels. Under normal conditions, the procedure described above provided for all of these requirements. Based on uranium assays of the secondary dissolution samples, we deduced that the combined action of the two dissolution stages resulted in at least 99.99% of the segment fuel being dissolved. By analyzing blend-tank solution from "blank" or "cleanup" dissolutions that were performed following dissolution of the last segment from each fuel rod in each dissolver, we determined that carryover of fuel in the dissolvers from one segment to the next did not exceed 0.01% of the previously dissolved segment. Since segments from a given rod were dissolved in order of increasing uranium content, this level of carryover was acceptably small. In total, fuel losses on the solution-transfer filter in each dissolver system and in the dissolver piping did not exceed 0.03% of the segment fuel during a normal run. This level of fuel loss did not jeopardize our ability to meet requirements on the various segment assays.

Most segment dissolutions performed during the EOL campaign were accomplished under these "normal" conditions. However, operational mishaps occurred during dissolutions involving 18 of the 152 LWBR fuel rod segments that were dissolved. The mishaps involved both mechanical failures, such as leaking transfer lines, and operator errors. Many of these mishaps were relatively innocuous in that their occurrence affected neither the magnitude of the assay results obtained for the corresponding segment nor the uncertainty associated with the results. The mishaps with eight of the LWBR fuel rod segments, however, involved a displacement of dissolver solution out of the dissolver system and required special recovery operations to account for the fuel in these displaced solutions.

Our approach to dealing with these mishaps was to collect, as completely as practical, the displaced solution and to assay the recovered material and include these assay results as part of the segment inventory. The quoted uncertainty in the assay result included additional terms that accounted for the additional measurements and also for potentially unrecovered material. The isotopy of uranium in the recovered material was used to verify that the material belonged to the particular segment of interest. In no case did the occurrence of a dissolution mishap impair our ability to meet project error requirements on uranium isotopic measurements or on fission-product assays. However, for five segments, uncertainty in the quantity of potentially unrecovered fuel in the displaced solutions increased the potential bias in the segment uranium assay results beyond the limits to allowable error. The probable errors in our assay results are discussed later.

Considering that 152 LWBR fuel rod segments were dissolved over the course of the 16-month EOL campaign, and that these dissolutions were carried out on a grueling 24-hour-per-day schedule involving some 14 different dissolver operators, the level of performance achieved in these operations appears extraordinarily good.

Chemical and Radiometric Analysis of Dissolver Solutions. Solution samples obtained during dissolution of each segment were analyzed to determine the isotopic composition of the uranium in the segment, the total uranium loading of the segment, and the amounts of selected fission products (^{137}Cs , ^{144}Ce , and ^{95}Zr) that the segment contained. The uranium isotopic composition was determined by analysis of single aliquots taken from each of two separate samples of the unspiked blend tank solution. Ratios of the uranium isotopes with mass numbers 234, 235, 236, and 238 to the isotope ^{233}U were measured by thermal ionization mass spectrometry (TIMS). Because of a potential isobaric interference by ^{232}Th in the TIMS measurement of ^{232}U , the ratio of ^{232}U to ^{233}U in each sample was measured by Alpha Pulse Height Analysis (APHA). Weight percent abundances of all these uranium isotopes in each segment were computed from the measured isotope ratios.

Although abundances of all the isotopes were reported for each segment, the fissile isotopes (^{233}U and ^{235}U) were of primary interest to BAPL. Project requirements permitted a potential bias in the sum of the abundances of these isotopes of 0.05% and a standard deviation of the sum of 0.08%, relative. For segments of low uranium loading (i.e., less than 0.2 g total U in the segment), these requirements were relaxed somewhat. Bias errors in these determinations arose from uncertainty in the NBS isotopic Standard Reference Material (SRM), U-500, used in calibrating the TIMS isotope ratio measurements and from error in the ^{232}U half-life used in calculations related to the APHA data. The magnitude of the percent relative bias associated with each individual weight percent abundance depends on the specific isotopic composition of the uranium sample. On the basis of a worst-case calculation consistent with the isotopy of the uranium in the EOL LWBR fuel rod segments, we determined that the maximum probable bias in the sum of the weight percent abundances of the fissile isotopes is only 0.016%, relative, well within the limit (0.05%) to allowable bias.

The standard deviation of each uranium isotopic result was estimated from uncertainties in the calibration of measurement systems, the making of measurements, and the introduction of traces of extraneous uranium into the samples prepared for analysis (i.e., the isotopic procedure blank). Every segment analysis

met requirements on the standard deviation of the fissile isotopes sum. In fact, except for two low-uranium segments, which showed relative standard deviations (RSD) between 0.08 and 0.09%, the RSD of the sum never exceeded 0.03%. Precision of the EOL isotopic measurements was, thus, quite satisfactory.

The uranium content of each dissolved segment was determined by combining data from measurements that were made on samples of solution that were taken from the dissolver and blend tank. For fuel-bearing segments, the total segment uranium was calculated in two virtually independent ways.

One way involved adding the quantity of uranium present in the spiked blend tank (SBT) solution to the (small) quantities of uranium that were removed from the dissolver system in the samples from the second dissolution (DS) and unspiked blend tank (UBT). These latter quantities were determined as the product of the weight of solution present in the samples and the weight concentration of uranium in the corresponding solution. The required concentrations were measured by MSID analysis of aliquots weighed from the samples and spiked with NBS SRM 993--a primarily ^{235}U isotopic and assay standard that we carefully cross-standardized against NBS 950a for this work (NBS 950a was designated the primary uranium assay standard for this project). Because uranium in the SBT represented the bulk (97% or more) of the segment uranium and because it was measured by MSID using the NBS 950a assay standard as spike, the total segment uranium result that was calculated from it was designated the 950a-MSID assay result.

The other way in which total uranium was calculated involved summing the uranium removed from the dissolver as DS samples and the uranium contained in the UBT before it was sampled. The unsampled UBT solution was weighed during dissolver operations so that the quantity of uranium that it contained could be computed as the product of this weight and the concentration that was measured, as described above, for the UBT samples. Because the uranium concentrations involved in this calculation were determined by MSID using NBS 993 as spike, the result of this calculation was designated the 993-MSID assay result. This result was considered to be a corroborative check on the uranium assay for each fuel-bearing segment, subordinate to the 950a-MSID result.

Corroboration of the 950a-MSID by the 993-MSID results was completely satisfactory. Percent relative differences between the two results for a given segment averaged only 0.0175% over the 135 fuel-bearing segments and showed a standard deviation of 0.089%. Both the magnitude and dispersion of these differences were consistent with expectations based on a comprehensive analysis of errors associated with each of the two MSID results.

Bias in the 950a-MSID total segment uranium results arose from two primary sources: (1) potential errors in the certified values of standards used in the assay measurements and (2) potential losses of fuel material during dissolver operations. The maximum probable measurement bias was 0.093%, relative, as estimated on the basis of the one-sigma uncertainties of the NBS 950a, NBS 993, and NBS U-500 standards that we employed. Under the conditions of normal dissolver operations, fuel losses did not exceed 0.03% of the segment total. Thus, by summing the fuel-loss and measurement bias contributions for segments dissolved under normal conditions, we determined a limit to probable bias in the

950a-MSID result of 0.12%, relative. The uranium assays for these segments thus met the project requirement that this bias be less than 0.15%, relative.

For those segments that were involved in dissolution mishaps, the fuel-loss bias estimates were higher than those for the normal dissolutions. As a result, potential bias in the 950a-MSID uranium assay for five segments exceeded the 0.15% limit, ranging from 0.18 to 0.34%. We were able to show, however, that potential bias in the sum of the uranium assay results for segments from any given LWBR fuel rod was less than or equal to 0.15%, in spite of one or more segments from the rod having a potential bias exceeding this value. Since the potential bias in this sum is no worse than if all the segment uranium assays had a 0.15% bias, we feel justified in concluding that recovery from the dissolver-operation mishaps was successful and that, on the whole, our uranium assays were of sufficient quality to meet LWBR-POB program needs.

The standard deviation of each 950a-MSID assay result depended on a number of segment-specific variables and was separately evaluated for each individual segment. Values for these estimated standard deviations ranged from 0.018 to 0.185%, relative. The value for only one of the 135 fuel-bearing segments exceeded the limit (0.15%) allowed by project requirements. This segment had a low uranium loading (0.22 g) that represented only a minor fraction of the uranium total (58.22 g) for the segments from this particular PFB-type rod. Its larger RSD (0.185%) was judged acceptable since it did not significantly affect the RSD of the sum of the segment uranium assay results for this fuel rod. Hence, the project requirement in regard to standard deviation of the segment uranium assays was satisfied. The high precision of the MSID assays was enabled, in large part, by our use of a unique, internal-standard procedure for measuring the needed uranium isotope ratios by TIMS. This procedure permitted measurement of a single isotope ratio with a standard deviation less than 0.01%, relative.

In addition to uranium assays on the fuel-bearing segments, ANL performed similar assays on the plenum segments from the LWBR fuel rods to screen the plenums for the presence of fuel material. Plenum segments were also assayed for thorium. The results of these plenum assays indicated the presence of fuel material in virtually all plenum segments, although the quantities of fuel involved were negligibly small in most cases. For three of the 17 fuel rods, the quantity of uranium in the dissolved plenum was sufficient to allow determination of the uranium isotopic composition. In each of these cases, the isotopy of the plenum uranium was found to agree closely with that of the fuel-bearing segment adjacent to the rod plenum, indicating that the origin of this fuel was the end of the fuel stack rather than carryover, on the shear or in the dissolver, of fuel from other rods.

The fission products ^{137}Cs , ^{144}Ce , and ^{95}Zr were determined for each fuel-bearing segment by gamma spectrometry on weighed aliquots of the UBT solution samples. A high-purity germanium detector and fully automated multichannel analyzer/data management system were used for this measurement. As was done for estimating the probable bias in ANL's uranium assay results for each segment, probable bias in the fission product determinations was estimated by summing contributions from the measurement and fuel-loss biases. Systematic errors in the measurements arose primarily from uncertainties in the certified values of standards used in calibrating the gamma-counting system, but also included minor contributions from potential errors in weighing the UBT solution and sample

aliquots. For ^{137}Cs , the largest maximum probable bias estimate had the value 0.43%, relative, well within the allowable limit of 0.5%, relative. For ^{144}Ce and ^{95}Zr , the largest values for the corresponding estimates occurred with the same segment and had identical magnitudes, 0.70%. For comparison, the limits to allowable bias in determination of these fission products were 2.0% for ^{144}Ce and 2.5% for ^{95}Zr . Clearly, project requirements regarding bias in the fission product determinations were satisfied.

Precision of the fission product determinations was limited, to a large extent, by counting statistics associated with measuring the appropriate gamma-peak intensities, but was also influenced by random errors in weighing the sample aliquots and the UBT solution and in calibrating the gamma spectrometer. Additional variance contributions had also to be included for those few segments that were involved in mishaps during dissolver operations. Separate estimates of the RSD for each fission product determination on each segment were generated by a computer program that compiled and processed the fission product data. Standard deviations of the ^{137}Cs determinations ranged from 0.25 to 0.56%, well within the 1.25% limit allowed by project requirements. The ^{144}Ce determinations also met requirements (allowable RSD of 2.0%), with estimated RSD values ranging from 0.49 to 1.10%.

For determination of ^{95}Zr , all error requirements were waived for measurements performed more than two years after shutdown of the LWBR and for segments of low burnup or low ^{95}Zr concentration. These waiver conditions translated to an exemption from requirements of all ^{95}Zr measurements performed after October 1984 and were established because ongoing radioactive decay of this short-lived nuclide (half-life of 64.02 d) made measuring it difficult. Only three rods were processed prior to the October 1984 waiver date. Among the segments from these rods, the RSD of the ^{95}Zr determination for only one segment failed to meet the 4.0% limit allowed by project requirements. Because this segment had a low concentration of ^{95}Zr , it was exempt from the requirement. Thus, all ^{95}Zr determinations for which the requirement was pertinent showed acceptable levels of precision; RSD values ranged from 1.21 to 3.76%.

We continued to report values for the ^{95}Zr in each segment processed after the waiver date, along with our best estimate of the uncertainty associated with the reported value. Early in the campaign, estimated RSD of the ^{95}Zr measurements was typically on the order of 10% or less. For nine segments processed late in the campaign, the ^{95}Zr gamma peaks were swamped by counts from ^{154}Eu , which has peaks at the same gamma energies as ^{95}Zr . In these cases, the presence of ^{95}Zr in the segments could not be verified and was reported as "not detectable." Detection limits based on counting statistics associated with the measurements were given in place of uncertainties for these determinations. Other segments processed late in the campaign showed much smaller $^{154}\text{Eu}/^{95}\text{Zr}$ ratios, similar to segments processed earlier, and had measurements with RSDs in the range of 3 to 30%.

Fission Gas Measurements. The amounts of fission gas (Xe and Kr) contained in each fuel rod plenum and in off-gas accumulated during each segment dissolution were determined on a best-effort basis by gas mass spectrometry. When combined with the mass of fission gas released during shearing of each fuel rod, as estimated from measurements with the in-line radiation monitor in the shear-cell ventilation system, this data provided a clear view of the distribution of

fission gas within each fuel rod. From the plenum-gas analyses, we found that the rod void volume contained between 0.01 and 0.15% of the fission gas in each fuel rod. Only 0.17 to 0.58% of the fission gas was released during shearing. The bulk of the fission gas (more than 99%) remained with the fuel material until it was released by dissolution of the fuel. We estimated that relative uncertainty in our fission gas determinations was on the order of $\pm 10\%$.

On a few occasions during processing of the LWBR fuel rods, dissolver off-gas samples were lost owing to mechanical problems such as pump failures and leaks in the off-gas retention systems. Rather than report no value for the fission gas contained in these segments, we reported values that were estimated by means of an apparent correlation that existed between the fission-gas content and ^{137}Cs content of segments belonging to a given rod. It was interesting to find that a single correlation line could be applied, within our estimated measurement errors, to all rods processed in the EOL campaign. This consistency of the fission-gas results relative to the ^{137}Cs determinations performed on the corresponding segments support the validity of the fission-gas values and the error limits that we assigned to them.

I. INTRODUCTION

The Light Water Breeder Reactor Proof-of-Breeding (LWBR-POB) Analytical Support Project at Argonne National Laboratory (ANL) is part of a national effort, sponsored by the Department of Energy-Division of Naval Reactors (DOE-NR), to develop the technology for breeding ^{233}U from ^{232}Th in a light-water reactor.¹ In the parent program, the Bettis Atomic Power Laboratory (BAPL), operated by Westinghouse Electric Corporation for DOE-NR, is carrying out a nondestructive assay of a statistical sample (500 fuel rods) from the end-of-life (EOL) core (~17,000 rods) of the LWBR which was operated at Shippingport, PA, from September 1977 to October 1982. The assay uses a BAPL-designed delayed-neutron device, the production irradiated fuel assay gauge (PIFAG), to measure the fissile loading of each rod in the sample. These measurements allow an estimate to be made of the EOL fissile inventory for the entire LWBR core. Breeding performance of the LWBR is measured through a comparison of the EOL fissile inventory with the known, beginning-of-life (BOL) fissile loading of the reactor.

ANL's role in the program (funded by the DOE-Division of LMFBR Fuel-Cycle Projects) was to carry out destructive analyses on a smaller number (17) of fuel rods after they had been assayed by BAPL with the PIFAG. The results of the ANL analyses will be used by BAPL to obtain correction and calibration factors related to the PIFAG measurements and to corroborate the results of the nondestructively determined fissile loadings. The ANL project was carried out in three phases. Phase I involved a pilot-scale program, which included the destructive analysis of experimental [Grid Rod In-Pile (GRIP-II)] irradiated rod sections (not irradiated in the LWBR) for calibration and validation of the prototype PIFAG device; this work was completed in 1981 and has been reported elsewhere.^{2,3} Phase II was directed toward the design, development, installation, and qualification of full-scale equipment, systems, and procedures for processing full-length (~3-m) rods from the LWBR EOL core; it culminated early in 1984 with the successful processing of a full-length GRIP-II rod (No. 79-440) that had been assayed by the PIFAG. The ANL results served to validate early measurements with the PIFAG. Phase III, designated the EOL campaign, used the facilities established in Phase II to conduct the destructive assays of 17 actual LWBR fuel rods that had been assayed with the PIFAG; the campaign was carried out in the period August 1984 to November 1985.

This report describes the equipment and methodology developed in Phase II, summarizes our experience in processing the EOL fuel rods (Phase III), and presents results that indicate the high quality of the individual measurements and analyses that were performed in the course of the work. Project error requirements governing the precision and accuracy to which each desired result was to be determined are shown to have been met or exceeded in all but a few cases. Even in those few cases where established project requirements were not met, our data analysis suggests that the quality of results is sufficient to meet the needs of the parent LWBR-POB program.

II. PROCESS REQUIREMENTS AND DESIGN CRITERIA

Process requirements and design criteria for the ANL work were defined on the basis of (1) the information required by BAPL for each fuel rod, (2) the precision and accuracy required for each measurement or analysis performed, (3) differences among the individual rods to be processed, and (4) the nature of the operations, which involved manipulations of highly radioactive materials and required hot-cell containment of much of the equipment. These factors are discussed individually below.

A. Required Measurements and Analyses

The ANL destructive assay of each LWBR fuel rod included a number of measurements and analyses. Physical measurements on the whole rod were made to determine rod length, weight, and surface temperature. The plenum section of the rod was punctured, and gases contained in the void volume of the rod were collected for analysis to determine the total moles of gas and the fraction of the total present as fission gas (Kr plus Xe); the isotopy of each fission gas was also measured. The rod was precisely sheared into segments according to a plan provided by BAPL; the actual location of each segment cut was identified relative to a specified reference point on the rod (the bottom end of the rod). After shearing, the length and weight of each segment were determined. Losses during shearing were evaluated by comparing the initial weight of the whole rod with the sum of the segment weights. Although not a requirement, fission gas released during shearing of each segment was also estimated.

Chemical and radiometric measurements were performed on the individual segments after each was dissolved. Gases released during the dissolution were collected and analyzed to determine the quantity of fission gas retained in the segment prior to dissolution. Samples of the solution containing the dissolved segment were analyzed to determine (1) the isotopic composition of the uranium in the segment (isotopes of mass 232, 233, 234, 235, 236 and 238), (2) the total uranium mass contained in the segment, and (3) the amounts of each of the three fission-product monitors, ^{137}Cs , ^{144}Ce , and ^{95}Zr .

B. Error Requirements and Specifications

Because of the small magnitude of the breeding gain expected from the LWBR (~1.5%) and the relatively small number of fuel rods to be processed destructively at ANL, restrictive limits to allowable error were established by BAPL for each of the measurements and analyses in the destructive assay. Most of the effort expended during Phase II of the project involved the development of equipment and procedures sufficient to meet these error requirements. Because of their importance in defining the scope of the ANL effort and in providing a standard against which the quality of the ANL results may be judged, these requirements are discussed in detail in this section.

Based on the expected needs of the parent LWBR-POB program, DOE-NR established error requirements for the EOL destructive assay in terms of allowable errors in calculated fuel-rod parameters that are used by BAPL in checking the PIFAG and in refining its calibration.⁴ These parametric requirements, designated the "DOE-NR Error Requirements," are presented in Table 1.

Table 1. Limits to Allowable Error in Fuel Rod
Parameters: DOE-NR Error Requirements

Parameter	Allowable Error for Segment Assay		
	Relative Bias	Relative Precision	Total Error
Location of Fuel Rod Segment	--	--	0.01 in.
Linear Fuel Density, g/in.	1.0%	1.0%	
U-Fissile Relative Loading, g/g of fuel		0.5% ^b	1.0%
U-Isotopic ^a (²³³ U + ²³⁵ U), % abundance		0.05% ^b	0.08%
FP Monitor Relative Loading, atoms/g of fuel:			
¹³⁷ Cesium	--	--	2.5%
¹⁴⁴ Cerium	--	--	5.0%
⁹⁵ Zirconium	--	--	7.5%

Parameter	Allowable Error for Whole Rods	
	Relative Bias	Relative Precision
U-Isotopic ^a (²³³ U + ²³⁵ U), % abund.	0.05% ^b	0.08%
U-Total ^c Rod Loading, g/rod	0.15% ^b	0.15%
U-Fissile ^c (²³³ U + ²³⁵ U) Rod Loading, g/rod	0.15%	0.15%

^aConcentration of other uranium isotopes (232, 234, 236, 238) is to be provided also.

^bMaximum bias values are relative to NBS SRM 950a (corroborated by NBS SRM 993) for the total uranium loading analyses, NBS SRM U-500 for ²³⁵U isotopic analyses, and BAPL ²³³UO₂(NO₃)₂•6H₂O reference material (²³³UNH) for ²³³U isotopic analyses. The frequency of use of these materials is not less than: NBS SRM 950a, each segment; NBS SRM 993, each segment; NBS SRM ⁵⁰⁰U, every fifth segment; ²³³UNH, every tenth segment.

^cU-total rod loading is defined as the total amount of uranium in the rod found in the course of dissolving and analyzing the segments into which the rod is cut. Similarly, U-fissile rod loading is defined as the total amount of ²³³U and ²³⁵U in the rod found in the course of dissolving and analyzing the segments into which the rod is cut. The sum of the U-total rod loading relative bias and the U-isotopic (²³³U + ²³⁵U) relative bias, which equals the U-fissile (²³³U + ²³⁵U) rod loading relative bias, shall not exceed 0.15%.

Because ANL would not actually measure all of the parameters listed in Table 1, but, instead, would make measurements and perform analyses to yield data from which the parameters would be calculated, BAPL provided a restatement of the limits to allowable error in terms of results obtained directly from the ANL work.⁴ These "Project Error Requirements" (see Table 2) were established by mutual agreement between BAPL and ANL to reflect realistically achievable performance levels for the ANL destructive assay and to ensure that the individual allowable measurement errors would propagate to the allowable parameter errors in Table 1. A listing⁴ of added specifications was included by BAPL among the project error requirements. The error requirements and specifications^{4,5} that were in effect at the time of qualification of the ANL systems are presented in Table 2 and in the ensuing paragraphs.

Specifications that were appended to the error requirements of Table 2 follow:

1. Each fuel rod is to be handled at ANL in such a way that the elevation of its plenum end does not descend below the horizontal.
2. From each segment fuel solution, ANL shall take archive samples adequate for a complete chemical re-analysis (U-isotopic, U-total, and fission product monitors). The archive samples are to be retained by ANL until BAPL gives permission for their disposal.
3. The fissile U-isotopic ($^{233}\text{U} + ^{235}\text{U}$) analysis shall be standardized against National Bureau of Standards (NBS) SRM U-500. The BAPL ^{233}UNH reference material will be used to assure that the bias requirements for $^{233}\text{U} + ^{235}\text{U}$ are not exceeded. The abundances of the remaining U-isotopes (232, 234, 236, 238) are also to be reported.
4. The U-total analysis shall be standardized against NBS SRM 950a (^{238}U). The sum of the U-isotopic relative bias and U-total relative bias shall not exceed 0.15%. To determine the acceptability of the U-total determination based upon NBS SRM 950a, the U-total determination based upon the NBS SRM 950a will be compared with the U-total determination based upon NBS SRM 993 (^{235}U). In making this comparison, the 993 result shall first be corrected for bias (95% probability level) with respect to the 950a U-total result.

After correction of the 993 U-total result for bias, so that the expected values of the 950a and 993 determinations are the same, the criterion for accepting the 950a U-total result is the following: the percent relative difference, $\% \bar{d}$, between the 950a U-total result and the bias-corrected 993 U-total result must lie within the 95% probability interval of the percent relative difference based on the allowable precision values, $\% \text{RSD}_{950a} = \% \text{RSD}_{993} = 0.15\%$. Thus, the critical value of the percent relative difference for acceptance of the 950a result is

$$\%d_{\text{crit}} = Z_{1-\alpha/2} ((\% \text{RSD}_{950a})^2 + (\% \text{RSD}_{993})^2)^{1/2} \quad (\text{II-1})$$

$$\%d_{\text{crit}} = 1.96 (0.15^2 + 0.15^2)^{1/2} = 0.416\% \quad (\text{II-2})$$

Table 2. Limits to Allowable Error in ANL Measurements and Analyses: Project Error Requirements

Analysis of Recovered Fuel Material	% Relative Bias	% Relative Standard Deviation
U-Isotopic ($^{233}\text{U} + ^{235}\text{U}$) (% abundance)	0.05, or (0.01 g/ $^T\text{U}_{\text{SEG}}$) ^a	0.08, or (0.01 g/ $^T\text{U}_{\text{SEG}}$) ^a
U-Total (g/seg)	0.15	0.15
^{137}Cs (atoms/seg)	0.5	1.25
^{144}Ce (atoms/seg)	2.0	2.0
^{95}Zr (atoms/seg)	2.5 ^b	4.0 ^b
Measurement	Bias	Standard Deviation
Rod Weight ^c , g	0.10	0.10
Rod Length, in.	0.001	0.010
Segment Weight ^c , g	0.001 if weight is <286 g, otherwise 0.01 g	0.005
Cladding Segment Length, in.	0.001	0.005
Fuel Segment Length, in.	0.005	0.015
Cladding Segment Boundary Location, in.	0.010 total	
Fuel Segment Boundary Location, in.	0.005 ^d	0.015

^aThe larger of the two requirements shall apply to any specific case;

^T U_{SEG} represents the total segment uranium, in units of grams.

^bThis requirement is waived for analyses performed more than two years after reactor shutdown or for low burn-up and low-concentration segments.

^cWeights are given as mass in air relative to 8.0 g/cm³ density standard weights.

^dNot including the average fuel shear-plane displacement, which will be corrected for. See Sec. V.B.1.a. for discussion of shear-plane displacement.

If %d between the 950a U-total result and the 993 U-total result obtained from a segment fuel solution does not satisfy this criterion, ANL shall repeat in duplicate both the 950a and the 993 U-total analyses on the archive samples retained from that segment fuel solution.

5. The frequency of use of the standards for the total uranium and isotopic uranium analyses shall be not less than: NBS SRM 950a, each segment; NBS SRM 993, each segment; NBS SRM U-500, every fifth segment; BAPL ^{233}U reference material, every tenth segment.

6. The ^{137}Cs analysis shall be standardized against the NBS SRM 4233 ^{137}Cs burn-up standard (atom concentration attribute). The ^{144}Ce and the ^{95}Zr analyses shall be standardized against standards of recognized high quality (e.g., radioactive standards of the Amersham Corp.).

7. The one-sigma uncertainty of a standard upon which a measurement or an analysis is based shall be taken as the bias contribution of that standard to the measurement or analysis.

8. In the pre-EOL developmental study, it shall be established experimentally that the conditions of sampling the EOL fuel solution result in negligible sampling error.

9. For the pre-EOL developmental study with the EOL shear, the following two requirements were specified:

- (a) The value of the overall recovery fraction of the fuel material from sheared LWBR-type fuel rods loaded with unirradiated thorium pellets shall be not less than 99.75%. (BAPL will determine, from the ANL data, the actual overall recovery fraction for each EOL rod sheared.)
- (b) An accurate RSD value of the segment fuel material recovery fraction shall be established in the shear development studies for calculating the segment assay parameters of all the EOL rods. The RSD of the segment fuel recovery fraction from these studies shall not exceed 0.25%. (The RSD of the segment fuel recovery fraction for an individual EOL rod cannot be obtained from the EOL shearing data.)

10. Weight and length measurements shall be calibrated against standards referable to nationally or internationally accepted standards.

C. Description of EOL LWBR Fuel Rods

The LWBR fuel rods processed at ANL were of four types, differing mainly in their LWBR core function and location. Each type is characterized by a distinct diameter. Nominal characteristics of the individual rod types are shown in Table 3.

All of the fuel rods, regardless of type, consisted of a fuel stack made up of ceramic pellets contained in Zircaloy-4 cladding. Depending on the type of rod and the location within a given rod, the ceramic pellets were composed (at BOL) of either thorium or a binary mixture of thorium and uranium ($\sim 98\% \text{ } ^{235}\text{U}$). The plenum (top) end of each rod also contained a stainless-steel (SS) sleeve and Inconel-X spring to restrain movement of the fuel stack. Individual rods of a given type were either top- or bottom-mounted, i.e., a threaded mounting tang was present on the corresponding Zircaloy end cap to facilitate mounting in the reactor. The other, nonmounting end cap on each top-mounted rod was hemispherical at its terminus, while bottom-mounted rods had a flat, chamfered end cap at their top end.

Table 3. Nominal Characteristics of the Four Types of LWBR Fuel Rods Processed at ANL

Rod Type	Nominal Diameter, in.	Nominal Length, in.	Nominal Weight, g
Seed	0.306	117	1120
Standard Blanket	0.572	118	4211
Power-Flattening Blanket	0.527	118	3560
Reflector	0.832	111	8530

D. Equipment Design Criteria

Besides having to provide measurements and analyses with the levels of precision and accuracy called for in the project error requirements and having to accommodate all the different rod types anticipated in the EOL destructive assay sample, ANL (in Phase II) had to develop equipment and methodology that fulfilled certain criteria imposed by the highly radioactive fuel rods. Hence, some of the equipment had to be designed for installation in shielded cells, to be remotely operable and maintainable, and to be fabricated from radiation-resistant components. In addition, transfer systems had to be devised so that they provided convenient movement of materials and samples between cells, but also maintained a high level of containment for the transferred materials. Radiation sensors were required in the hot-cell ventilation exhaust stacks to monitor accidental or intentional releases of radioactive gases, such as ^{85}Kr or ^{220}Rn , given off during processing of the fuel rods.

The requirement that the bulk of the work be performed in a shielded environment affected not only the design of the hardware developed for the project, but also the methodology and procedures developed for carrying out the work. Automated operation was given preference wherever practical to enhance productivity and to reduce the number of operator manipulations that had to be performed with master/slave (M/S) manipulators. The methodology for taking and preparing samples for the chemical and radiometric measurements made on the dissolved fuel rod segments was chosen to minimize the need for quantitative transfers of solution and quantitative recovery of sample materials during chemical processing and purification steps.

Also of major importance in establishing the design of equipment and methodology for the EOL destructive assay was the need to manage the large amounts of data produced during the processing of each rod. A data-management system had to be established to provide for error-free data acquisition, completeness of data sets, reliable data storage and accessibility, and the timely reporting of results. Complete data sets were ensured by adopting computer-controlled procedures for those operations where irreversible changes were made in the materials processed, e.g., shearing of the rods or dissolution of the rod segments. Computer control, in these cases, provided confidence that the

sequence of steps involved in each operation would be carried out in the same order each time and that recording of a critical data item would not be overlooked. The computer system developed for controlling the execution of procedures also provided the tools needed to fulfill the other data-management needs of the project.

III. FACILITIES AND EQUIPMENT

The destructive assay of the LWBR fuel rods processed at ANL was carried out with equipment developed and qualified specifically for use by the LWBR-POB project in ANL facilities. Equipment used in handling the highly radioactive materials involved in processing the rods was installed in hot cells within the Shielded Cell Facility of ANL's Chemistry Division (CHM). Other equipment was located in facilities associated with ANL's Analytical Chemistry Laboratory (ACL), which performed chemical and radiometric analysis of solution and gas samples generated during the processing of each rod. The ACL also prepared reagents and chemical standards for use in project operations. Process and analytical instrumentation was interfaced to a fully integrated computer system, which was used for process control and data management.

The facilities, processing and support equipment, and the computer system are described in this section.

A. Hot Cells

1. General

The Shielded Cell Facility at CHM is a twelve-cell complex constructed on two levels (Fig. 1). The cells on each level are arranged around a shielded central corridor having tracks and turntables used in the operation of a radio-controlled, battery-powered cart (MULE), which facilitates movement of materials among the individual cells; a remotely operable elevator within the shielded corridor permits transport of the MULE and materials between levels. A loading dock equipped with a 10-ton crane and cask-unloading area connects directly to the corridor on the lower level.

The hot cells in the facility are of three types (megacurie, kilocurie, and analytical), which differ according to cell dimensions, level of shielding, and ventilation provisions. The four megacurie cells are located on the lower level of the complex and have the greatest amount of shielding, i.e., 48-in. thick walls constructed of magnetite concrete; these are also the largest of the cells. The kilocurie cells also have magnetite concrete walls (28-in. thick), while the analytical cells, which are smaller, have shielding walls (28-in. thick) of ordinary concrete; these cells are located on the upper level. Illumination in each cell is provided by mercury-vapor lamps with additional tungsten-halogen emergency lamps for backup. Each cell and several locations along the shielded corridor on each level are equipped with one or more work stations, each consisting of a zinc-bromide-filled viewing window and a pair of M/S manipulators, many of which have extended-reach capability. Top-suspended, concrete shielding doors isolate each cell from the adjoining corridor. Shielded and ventilated storage tubes are provided in the floors of some megacurie cells and the lower-level corridor.

Equipment for the EOL destructive assay was installed in five separate cells within the complex: three megacurie cells and two kilocurie cells. Because rods were received in both single and multiple-rod shipments but only one rod was processed at a time, one of the megacurie cells was used exclusively for storing LWBR fuel rods awaiting processing. This cell was outfitted with simple fuel-rod storage racks constructed of angle iron and designed to provide uniform support along the length of each rod, as well as the required elevation of the

Fig. 1. Shielded Cell Facility. (All work involving LWBR fuel rods is carried out in five of the twelve cells.)

rod's plenum end. The other four cells were outfitted with the equipment required for the actual EOL operations. One megacurie cell, designated the Full-Scale Shear Facility (FSSF), contained equipment used in obtaining physical measurements on whole fuel rods and for shearing each rod into precisely characterized segments. Another megacurie cell contained the Dual Dissolver System (DDS), which comprised two fully independent dissolver units and ancillary equipment for the quantitative dissolution of the individual sheared

segments. The kilocurie cells were equipped for (1) preparing samples from the dissolution of segments in the DDS for subsequent analysis (the Analytical Hot Cell) and (2) for converting the bulk of the dissolver solutions to a cemented waste for disposal (the Waste Treatment Facility). All four cells used for project operations shared a number of common features, which are described immediately below. Each separate hot cell also contained specialized equipment for carrying out the operations specific to a given cell; this equipment is described in the following sections for each individual cell.

2. Features Common to All Project Hot Cells

a. Work Table and Alpha-Containment Barrier

An alpha-containment structure was fabricated for each cell used in project operations to minimize the potential for spreading contamination into the corridor areas. Each containment structure consists of an elevated work table and wall and ceiling assemblies fabricated of aluminum or SS sheets and Lexan (product of General Electric Co.) panels bolted to a metal frame. The frame mates with two of the cell walls to complete the enclosure. Joints where the sheets or panels meet the metal frame were gasketed with rubber sheeting and sealed with silicone sealant. Individual components of the enclosures were sized and selected, insofar as practical, to facilitate disassembly, including the cutting of structural components during dismantling and disposal of the facilities at the end of the project. The actual cell walls were coated with ordinary vinyl wallpaper to prevent contamination of the concrete surfaces and, hence, to facilitate decontamination of the cells during the post-EOL cleanup.

b. Transfer Systems

To provide for movement of materials and equipment into and out of each cell across the alpha barrier, several types of transfer system were installed. One system was a double-door transfer hatch, constructed of Lexan on an angle-iron frame and a SS slide tray that extended into or out of the hatch to provide easy access to items being moved. The transfer hatch was primarily intended for use in moving tools and small equipment items into the alpha enclosure. Another transfer system consisted of bagout ports incorporated into the panels of the alpha barrier; these were used for removing waste and contaminated equipment from the hot cell. Commercial heat sealers, modified for convenient, fully remote operation, were installed in the corridor adjacent to the bagout stations of the cells other than the Analytical Hot Cell. The analytical cell waste and equipment were generally of sufficiently low activity that fully remote "bagging-out" was not required at this cell.

For the routine movement of materials and samples among the various hot cells (e.g., movement of sheared fuel rod segments or dissolver solution samples), a unique transfer system was devised. This system has pneumatically powered slide trays and an electric jack to position a positively sealed SS transfer can against a hatch assembly located in the surface of the hot-cell work table. When the cell operator opens the hatch assembly, the lid of the can is connected to the underside of the hatch door, with a gasketed seal, before the bayonet closure of the can is disengaged and the can is opened. The top lip portion of the can is sealed by the jacking mechanism against the base of the in-cell hatch assembly. Thus, when the hatch and can are open, only the interior of the transfer can is exposed to the contaminated cell environment. ANL was

granted a U.S. patent on the design of this can-out transfer system. The sealed cans were transported between cells by means of the MULE.

c. Ventilation Exhaust-Stack Radiation Monitors

Each cell is equipped with its own ventilation blowers and exhaust stacks, thereby allowing a wide variety of operations to be carried out simultaneously in the Facility. Before stack release, all exhausted air passes through a series of HEPA filters to retain potentially radioactive particulate matter. For the cells used in POB project operations, radiation sensors were installed in the ventilation exhaust stacks in cooperation with the Health Physics section of ANL's Division of Occupational Health and Safety (OHS). These sensors are part of a computerized stack-monitoring system developed by OHS for the project, which permits continuous, real-time measurement of a variety of radioactive gases that might be present in each cell's exhaust; these include ^{85}Kr , ^{220}Rn , and ^{129}I . Signals from the stack monitors may be integrated to provide values for the total release of any of these gaseous nuclides during any particular project operation.

d. Remotely Operated Electronic Balances

Many of the EOL hot-cell operations involved weighings of objects such as fuel rods, segments, or solutions. Thus, each of the operations cells was equipped with one or more top-loading electronic balances, each having a capacity and resolution consistent with the weighings for which it was used. All of the balances used in the project were from the Mettler "K"-series (Mettler Instrument Co., Hightstown, NJ), which covered the entire operating range from 30-g capacity and 0.01-mg resolution (Model HK-60) up to 60-kg capacity and 1-g resolution (Model PK-60).

The Electronics Shop of ANL's Chemical Technology Division (CMT) modified each of the balances installed in the hot cells to separate radiation-sensitive electronic components from the radiation-resistant load-cell. Before this modification, the radiation sensitivity of the balances was identified with components of the "digital" and "interface" printed circuit boards of each unit by means of tests done in the Gamma (^{60}Co) Irradiation Test Facility of the CHM Division. The sensitive components were installed in separate out-of-cell cabinets, which were connected by cables to the in-cell units. In initial testing, some of the balances showed poor linearity and precision after the electronics were made remote from the load cell. This problem was overcome by relocating one of the printed circuit boards (the "analog" board) in a lead-shielded compartment within the load-cell cabinet (i.e., the in-cell portion of the remotely operated balance) and then connecting the two cabinets of each balance with a special cable obtained from the manufacturer. This cable had a construction and shielding arrangement that provided minimal capacitive pickup and cross-talk between transferred signals. Testing of each remotely operated balance after installation within the hot-cell demonstrated performance on a par with manufacturer's specifications for the original unit in terms of precision, accuracy, and linearity of the instrument.

For each in-cell balance with a resolution of 10 mg or less (i.e., those balances whose readings were particularly sensitive to air currents, drafts, etc.), the in-cell cabinet was installed in a SS well set into the floor of the cell

work table; each balance well was equipped with a Plexiglas dust-and-draft shield that could be pneumatically opened and closed from a foot switch at the cell work station.

Each in-cell balance also had a set of calibration and test weights for use in quality assurance operations. For balances with a capacity of 2 kg or less, the in-cell standard weights were Class S standards procured from Mettler Instrument Co. and verified by comparison with standards that were individually calibrated by the U.S. National Bureau of Standards (NBS). Because commercial standard weights of mass in excess of 2 kg could not be conveniently handled remotely with manipulators, the calibration and test weights for the larger in-cell balances were designed and fabricated at ANL in 5-kg units; each 5-kg weight was turned from SS and polished. The shape of the weight included a narrow-diameter section that could be gripped by a manipulator. The mass of each weight was adjusted to $5 \text{ kg} \pm 0.1 \text{ g}$ (relative to NBS-calibrated standards) by adding SS shot to a cavity in the weight; the cavity was closed with a threaded eyebolt that constituted part of the weight's mass. Groups of weights designated for a specific balance were adjusted together such that the total mass of the group also agreed with the nominal total mass within 0.1 g. The standard weights were stored in closed Plexiglas boxes located near the balances in each cell to protect them from dust, falling objects, and potentially corrosive environments within the cell.

3. The Full-Scale Shear Facility (FSSF)

The FSSF contained equipment for handling and performing physical measurements on whole LWBR fuel rods and for shearing each rod into precisely characterized segments for subsequent dissolution and analysis. A special rod-entry transfer port was included in the alpha-containment barrier of this cell to permit unloading of each rod from its secondary shipping container directly into the hot cell. Major equipment items located in the FSSF included three remotely operated electronic balances, a pneumatically powered portable bolt cutter for removing the mounting tang from each rod, a rod-puncture and gas-collection system, and a computer-controlled, automated hydraulic shear having a feed/positioning system capable of positioning the rod for cutting within 0.001 in. of a desired location relative to the reference end of the rod. Each of these items is described separately below.

a. Weighing Systems

The three balances in the FSSF were used in weighing the whole fuel rods and the segments sheared from each rod; two different balances were required for segment weighings to meet requirements over the entire range of anticipated segment weights. The fuel-rod balance was part of a rod-weighing system that was designed to provide the requisite high accuracy and precision, along with nearly full-length rod support and ease of operator manipulation. The two segment-weighing balances were located in covered balance wells as described in Section III.A.2.d.; they included a Model PK-300 instrument (300-g capacity, 0.001-g resolution) and a Model PK-2000 (2-kg capacity, 0.01-g resolution), both remotely operated.

The fuel-rod weighing system comprised a remotely operated Model PK-16 balance (16-kg capacity, 0.1-g resolution) equipped with a specially designed Plexiglas draft shield, a 10-ft-long aluminum trough with detachable carrying hooks, and two bearing yokes, one on each side of the balance. The

yokes could be raised or lowered by air-powered pistons controlled from outside the hot cell. The draft shield was slotted to facilitate placement of the trough on the balance pan. The need for a draft shield on this system was identified during early testing, which showed that the strong airflow in the FSSF cell adversely affected the precision obtained with the unshielded balance.

For weighing, the fuel rod was rolled from the shear bed onto the aluminum trough, and the trough was positioned such that one of its ends rested in each of the bearing yokes. The yokes were then lowered by means of the air pistons, allowing the trough (with the rod in place) to lay gently on the PK-16 balance pan; the right-side yoke, which bore the plenum end of the rod, was set up to descend more slowly than the left yoke so that the plenum would not fall below the horizontal, as per requirements. In raising the load from the balance pan, the right-side yoke moved more quickly than the left for the same reason. The weighing system allowed for repetitive weighings of the trough or the trough and rod combined. This was accomplished by completely removing the object from the balance pan and replacing it for each weighing and did not require multiple manual operations. The complete removal and replacement of an object during repetitive weighings give more meaningful weighing statistics than would simple multiple readings with the object on the balance pan.

b. Pneumatic Bolt Cutter for Tang Removal

The presence of the mounting tang on each fuel rod interfered with indexing the rod mounting end against reference stops on the shear bed during measurement of rod length and shearing of the rod. Therefore, a commercial pneumatic bolt cutter, modified for remote operation and handling, was used in removing the tang. The bolt cutter was mounted vertically in a metal frame in such a way that it could be raised and lowered to engage the tang in its jaws, while the rod rested flatly in a support fixture on the FSSF cell floor; the system accommodated all four rod sizes. With this tool, the mounting tang could be sheared from the Zircaloy endcap on the rod such that only a 1/8-in. protrusion on the endcap remained.

c. Rod-Puncture and Gas-Collection Station

The FSSF system for puncturing fuel rods and collecting the released gases was initially planned to satisfy processing needs but was fabricated to supply analytical information on each rod as well. A potential for high gas pressure in the void volume of EOL rods from the LWBR prompted plans to puncture each rod in a controlled manner prior to shearing so that the plenum gases would not be suddenly released during the first shear cut through the rod. The planned puncture also provided for collection of the plenum gas. Subsequently, the gas was slowly released to the cell ventilation air in a manner consistent with environmental limits for stack emission. By providing a means to measure the quantity of gas collected and to take samples of the gas for analysis, we could obtain information on the amount and composition of gas in the rod void volume with little added effort.

The rod-puncture rig comprised a steel yoke, which aligned a drive screw with the head of a tool-steel piercing pin in a saddle-type puncturing die, a gas-handling vacuum system consisting of two calibrated expansion volumes, and a gas sampling station, all connected in series. The yoke is designed to

accept any of four sizes of puncturing dies, corresponding to the four types of LWBR rods. The puncturing dies are fabricated in two sections, which are bolted together around the fuel rod to form a gas-tight, gasketed seal against the outer surface of the rod; the piercing pin moves through a sliding O-ring seal in a channel normal to the rod surface. Each expansion volume consists of a SS cylinder and connected piping that ties the gas-handling system to the puncture die and the parts of the gas-handling system to each other; the expansion volumes have separate capacitive manometers and thermocouples for measuring the pressure and temperature of the collected gas. The gas-sampling station is located in a hood attached to one of the outer walls of the hot cell and contains the piping needed to connect 150-cm³ SS cylinders, used for sampling the gas, to the system; the cylinders are fitted with plug-type valves for closure and self-sealing quick-disconnect fittings for connection to the gas system.

The two calibrated expansion volumes in the gas collection system allow an estimate to be made of the rod void volume. This value is needed to establish the quantity of gas released from the rod, as shown in Section IV.D.4, where calculation of the analytical results obtained from the rod-puncture operations is described.

d. The Full-Scale Shear (FSS)

The principal equipment item in the FSSF is the full-scale shear (FSS) itself. It consists of a hydraulically driven, vertically mounted shear, a sheared-fuel collection and canning station, and a digital-controlled feed/measurement system. Each subsystem on the FSS is designed to meet project requirements for precise location of shear cuts and efficient collection of sheared materials. The in-cell portion of the FSS is shown schematically in Fig. 2. Other shear controls and instrumentation, including the hydraulic power unit, are located outside the hot cell. Control and measurement instrumentation on the FSS is fully interfaced to the project computer system (see Section III.C) and provides automated shear operation and data collection. Automatic operation of the FSS is controlled from a video display terminal located at one of the hot-cell work stations. Construction of the FSS is modular, allowing remote replacement of individual components with minimal disturbance of other parts of the system.

The major subsystems of the FSS (i.e., the base, the cutting head, fuel collection station, and feed system) are described in the following subsections. Automated operation of the FSS is described in Section IV.D; performance of the FSS during characterization and qualification testing and during the EOL campaign is discussed in Sections V.A and B.

(1) The Shear Base

The base of the FSS is built around an upright wide-flange beam, which provides a rigid support platform. The shear assembly is aligned with and bolted to this beam at one end. The feed system components are mounted atop the beam's flange. Welded pads along the beam's sides provide bolt-down locations for the segment collection station, rod positioners, and other components.

Fig. 2. Schematic of Full-Scale Shear

(2) The FSS Cutting Head Assembly

The cutting head or shear tooling on the FSS consists of a tool-steel blade guided within a close-fitting housing, as illustrated in Fig. 3. Separate tooling, specifically sized to accommodate an individual rod diameter, is used for each type of LWBR fuel rod; the tooling for all four rod sizes is, however, similar in design and function. The shear blade has a rectangular cross section, nominally 1-in. wide by 0.75-in. deep, and is made of type M-42 tool steel hardened to Rockwell C66. The shear-blade housing, made of D-2 tool steel, guides both the blade and the fuel rod. Clearance between the blade and housing is small (0.0015 in.) to minimize errors in shearing cut location and to reduce losses of pulverized fuel.

The fuel rod enters the shear housing through a replaceable insert piece. The inside bore of the insert is closely matched to the diameter of the fuel rod to prevent fuel losses through the rod-to-insert diametric clearance. As a further precaution, to accommodate potential variations in irradiated rod diameters, an O-ring seal is incorporated into the insert assembly. The seal is made by compressing a flexible O-ring around the fuel rod during each shear cut; compression of the O-ring is automatically relaxed as the rod is fed into the shear, thereby preventing abrasion of the O-ring.

Each shear housing also has a rear fuel-rod-exit port, which allows the rod to be fed entirely through the housing and also allows use of a fixed stop located at this end of the machine (hereafter referred to as the "forward stop"). The forward stop has a recess in the form of a vertical slot machined into it to accommodate the mounting-tang protrusion on top-mounted rods; the tang protrusion enters the recess and thereby permits the shoulder

FULL-SCALE SHEAR BLADE/HOUSING ASSEMBLY

Fig. 3. Cutting Head of Full-Scale Shear (Exploded View)

of the top end cap on the rod to contact the face of the stop. This feature was included so that the tang protrusion would not be included in the rod length, as per BAPL's specification. The slot was contoured, wider at its top end than at the bottom, so that the correspondingly larger diameter mounting tang on larger diameter (e.g., reflector) rods would enter the recess and yet the narrow end-cap shoulders of the small diameter (e.g., seed) rods would mate securely with the face of the stop. This "extension" of the feed table plays an important role in making fuel rod length measurements with the FSS feed system, as described in Section IV.D.5. During shearing operations, the exit port in the housing is sealed with a tight-fitting bore plug.

The shear blade couples directly to a 4-in. bore, 1.5-in. stroke, hydraulic cylinder mounted on a support plate above the shear tooling. The cylinder connects to the out-of-cell hydraulic power unit via two (1/2-in. ID) flexible hoses. The hydraulic system provides a shearing force of up to 12,000 kg and shearing speed up to 50 in./s. The hydraulic power unit has an accumulator-unloading circuit in which high-pressure fluid is delivered to the cylinder by expansion of a hydropneumatic accumulator. After a shear cut, the accumulator is recharged by a small pump, and the shear blade is retracted for the next cut. Total cycle time (from shear stroke to shear stroke) is about 30 s. The hydraulic fluid (Pydraul 50E, Monsanto Co., St. Louis, MO) is water based.

A linear variable differential transformer (LVDT), incorporated into the hydraulic cylinder coupling, facilitates automatic acquisition of information on blade position and speed. Safety interlocks, to prevent inadvertent shear operation, are actuated by this sensor.

(3) Segment Collection and Canning Station

The segment collection and canning station is a pneumatically operated, rotating turret assembly with two positions designed for separate purposes. One position on the turret is designed to hold the aluminum sample can in which each sheared segment is collected and to raise the can against the underside of the shear housing, which is machined to a smooth finish so that there is a tight seal to the top of the can. The other turret position contains a blade-cleaning device in which a small, replaceable nylon disk (brush) may be raised and rotated against the underside of the shear blade and housing to remove residual fuel and cladding particles. This device helped to reduce fuel carryover between adjacent segments sheared from each fuel rod. The station also includes a demountable capping mechanism used to seal the aluminum cans.

The collection station is entirely remotely operable and contains sensors to prevent operation of the shear if a sample can is not in position to receive the fuel. If the station fails, it is replaced as a unit.

The sample collection cans were fabricated (impact-extruded) from type 1100-Al slugs. The cans, nominally 1.5-in. diameter and either 3 or 6 in. tall, were subsequently chemically milled to reduce the quantity of aluminum charged to the dissolver with each segment because of solubility considerations; the weight of aluminum in the dissolver plays a role in determining the minimum volume of dissolvent required for segment dissolution, and lighter cans allowed smaller volumes to be used.

(4) Feed/Measurement System

The feed system on the FSS is a high-precision positioning and measurement system used for the determination of fuel rod length, segment boundary locations, and segment lengths during processing of the EOL LWBR fuel rods. This system centers around a precision-machined ball screw, which drives a carriage upon accurately aligned linear bearings (carriage ways) affixed to the FSS base. Cylindrical rollers extending the length of the feed system support the fuel rod while measurements are made and the rod is being cut. Nine vertically mounted pneumatic hold-down clamps keep the rod centered on these rollers and prevent rod bowing, which would impact these operations. The hold-down clamps automatically retract as the carriage passes, actuated by the carriage-striking limit switches mounted along the front side of the machine. Three of the hold-down clamps (Nos. 1, 2 and 7) have resistance temperature sensors (RTDs) affixed to them such that rod surface temperature is obtained when the positioners are lowered. These RTDs are also used to measure the average temperature of a gauge rod used in establishing reference points on the feed system (see below).

Length and position information from the shear feed system is not obtained directly, but rather is computed as the difference between a set of transducer readings obtained with the carriage positioned (1) at a specific reference point (established using a gauge rod of accurately known length) and (2) then at the bottom end of the fuel rod. One of the transducers involved in the feed system measurements, the absolute shaft transducer (AST), indicates the position of the carriage on the feed table relative to the plane of the shear blade cutting edge; another, the rod-end locator (REL), indicates the position of the end of the fuel or gauge rod relative to the reference plane of the carriage.

Carriage movement is accomplished by rotation of the ball screw, which advances a preloaded nut assembly that is bolted to the carriage. The screw threads are machined such that each revolution provides a carriage travel of 0.2000 in. at a specific reference temperature (19.56°C). The ball screw is driven by a high-response, direct-current, permanent-magnet-type servo motor coupled to a digitally controlled, pulse-type, motor driver in a closed-loop feedback system ("Index-Syn," DC Motor and Closed Loop Positioning System, Control Systems Research, Inc., Pittsburgh, PA). The motor driver produces a rotation of 1/200 revolution per stepping pulse so that, at the ball screw reference temperature, each pulse corresponds to a movement by the carriage of 0.001 in.

The AST is a rotary transducer, mounted at the opposite end of the ball screw from the driving motor, and is independent of the driving system. The AST is a gear-driven, digital-readout counter (Northern Precision Laboratories, Inc., Part #801537-0, Fairfield, NJ) that accumulates 200 counts per revolution of the ball screw. Thus, at the ball-screw reference temperature, each count on the AST also indicates a carriage movement of 0.001 in. The digital readout on the AST is calibrated to read directly in inches. That is to say, the decimal point in the display is positioned so that an accumulation of 1000 counts results in a reading of 1.000. We emphasize that, although the motor driver and the AST are calibrated to move and to detect movement in 0.001-in. increments, the two are completely independent of each other. By design, the motor driver with its feedback control is generally the more reliable system, allowing the carriage to be positioned with better precision and accuracy (i.e., nearer to exactly 0.001-in. steps) than the AST position indicator. Nevertheless, the AST is used as the primary transducer for measuring carriage position for two reasons. First, the AST measures ball-screw rotation directly, while the motor driver measures rotation indirectly by counting pulses to the driving motor; the AST is thus less prone to errors due to mechanical malfunction of the driver. Second, the AST retains its reference-point setting even when the unit is shut down; in contrast, the motor driver measures displacement only and loses its reading in the event of a power loss. Therefore, from the standpoint of recovery-mode operations following a system failure during shearing, the AST is more reliable.

At temperatures other than the reference temperature for the feed system, thermal expansion of the machine base (including the ball screw) causes a change in the linear equivalent of the AST readout. To take into account this effect, the computer monitors temperature at the machine base using three RTDs attached to this base and automatically makes corrections for thermal expansion to the AST readout.

In the use of the AST, the reference setting, i.e., the zero reading, is set to correspond to the plane of the cutting edge of the shear blade (the "shear plane"). This setting is made with a nominally 35-in. gauge rod having an accurately known (± 0.0001 in.) length traceable to NBS standards. To establish this setting, the gauge rod is positioned on the shear bed such that one end of the rod contacts the lowered shear blade. Then, the carriage is advanced until the other end of the gauge rod moves the REL to the midrange of its travel (a REL reading of zero). The AST reading is adjusted so that its temperature-corrected value corresponds to the temperature-corrected gauge rod

length to within 0.0005 in. The reference point on the REL is taken to correspond to any residual difference (<0.001 in.) that might exist between the temperature-corrected AST value and the gauge rod length.

The REL is a LVDT mounted directly to the feed-system carriage, with its movable sensing shaft pointed in the direction of carriage travel. The REL is sensitive to a displacement of its shaft end as small as 25×10^{-6} in. and can measure such displacement over the entire LVDT operating range of ± 0.05 in. The REL serves several purposes. As mentioned above, it defines a reference position on the carriage for which the temperature-corrected AST value provides a true measure of distance from the shear plane. By measuring the amount by which the end of a rod is displaced from this reference position, the REL becomes an important part of the FSS measurement system, adding a finely divided scale that complements the "coarse" AST readout. The latter has limited length resolution because it responds only in 0.001-in. increments. By monitoring both the AST and REL readouts, one can obtain rod-end positions readable (not necessarily reliable) to 0.00001 in.

The end of the REL sensing shaft is machined to accept interchangeable adapters designed to seat flatly against the ends of rods having different diameters and endcap configurations. In particular, special adapters were required to seat against the squared-shoulder endcaps of bottom-mounted LWBR fuel rods that had a protrusion from the previously removed mounting tang; the reference point for shearing these rods was the square shoulder of the endcap. Changing the adapter on the end of the REL or replacing the shear tooling changes the reference points on the feed system. As standard practice, however, the reference points were reestablished using the gauge rod prior to each use of the system, regardless of whether or not the REL adapters or shear tooling had been changed since the last use.

A clamping assembly on the front end of the carriage holds the rod end in position against the REL during shearing. The jaws of the clamping assembly are spring-loaded to (1) allow up to 0.050-in. movement of the rod end in the event of obstructed rod travel, thereby avoiding damage to a fuel rod, and (2) return the clamp jaws to a specific, positive-stop position relative to the carriage when force on the rod is relieved. A separate LVDT monitors the position of these jaws.

Use of the FSS feed/measurement system for determining fuel rod lengths, segment boundary locations, and segment lengths is described in Section IV.D.

4. The Dual Dissolver System (DDS) Facility

Two complete and virtually independent dissolver systems were installed in one of the megacurie cells for use in dissolving the individual segments sheared from each fuel rod. The cell that housed the dual dissolver system (DDS) contained three work stations. Two of these were dedicated to operation of the dissolver units, one for dissolver system 1 (DS1), and the other for dissolver system 2 (DS2). The third work station served as a general work area for handling transfers of equipment, samples, and waste into and out of the cell's alpha enclosure; for cleaning, assembling, and testing reusable dissolver-system components between runs; and for performing remote maintenance and repairs on miscellaneous dissolver equipment. A remotely operable, 1-ton bridge crane in the

cell was used to move equipment between work stations and to perform specific operations with the dissolvers.

Each dissolver system consisted of (1) a tantalum dissolver vessel; (2) an off-gas handling system, which provided for dissolver pressure control as well as for collection and sampling of gases released during each segment dissolution; (3) a solution transfer system, including an in-line filter assembly used for removing and sampling solution from the dissolver vessel; (4) a vacuum transfer system used to implement solution transfers from the dissolver vessel to the blend tank (BT); and (5) a BT assembly into which all dissolver solutions and rinses from an individual segment were transferred so they could be mixed, weighed, and sampled for analysis. The two dissolver systems shared a common reagent-charging system, part of which was located in a fume hood attached to one of the outer cell walls.

Ancillary equipment in the dissolver cell included three remotely operated electronic balances for weighing segments and solution samples (these were in addition to the two balances included in the BT assemblies at the dissolvers), a lead-lined container for storing segments awaiting dissolution, and a liquid-waste storage system used to hold the fuel-bearing waste solution for subsequent conversion to a solid form and disposal. Besides these major items, the cell was also equipped with a variety of tools, jigs, and fixtures for undertaking mechanical operations within the cell, all of which were done with M/S manipulators.

Almost all of the control and measurement components in the DDS facility (e.g., valves, pressure transducers, thermocouples, pumps, heaters, etc.) were interfaced to the project computer system to allow computer control of operations and automated process monitoring and data acquisition. Two video display terminals were located at each dissolver work station. One was used for process control, while the other displayed graphical information regarding process variables such as temperatures, pressures, etc., on an operator selection basis during a dissolver run.

The major subsystems within the DDS are described individually below. Operations involving these subsystems are discussed in Section IV.E.

a. The Dissolver System

The two dissolvers in the DDS were installed in a mirror-image arrangement in the DDS cell. They were constructed of identical components, but shared no common parts so that they could be operated simultaneously and independently of each other.

Each dissolver system centered around a pressurized-reflux dissolver vessel with 30-L capacity, illustrated in Fig. 4. Each vessel consisted of a tantalum-alloy primary vessel that was close-fitted within an SS secondary safety shell; an integral water-cooled condenser was built into the upper section of the secondary vessel. Cooling water for the condenser was delivered by a closed-loop circulating system that included a separate coolant-supply pump in the primary loop for each of the two dissolvers, and a third pump in the shared secondary loop. The cooling-system heat exchangers were outside the cell; a radiation sensor built into the out-of-cell piping monitored gamma activity in the primary-loop water for early detection of any breach of the system that might

SEGMENT DISSOLUTION

- FLUORIDE-CATALYZED HNO_3
- 195°C , 125 PSIG
- PRIMARY DISSOLUTION – 4 HR
- SECONDARY DISSOLUTION – 3 HR

SOLUTION SAMPLING/SPIKING

- SOLUTIONS BLENDED/WEIGHED (± 0.7 g)
- TEN SAMPLES DRAWN AT 3 STAGES IN PROCESS
- BLEND TANK SOLUTION SPIKED WITH NBS STANDARD URANIUM SAMPLE
- ARCHIVAL SAMPLES STORED FOR ONE YEAR

Fig. 4. Dissolver Vessel and Blend Tank System

occur. With both dissolvers operating at full capacity, a coolant flow of 10 L/min (2.7 g/min) at each dissolver and a typical input/output coolant temperature differential of 10 to 12°C were achieved with this system.

Each dissolver was heated by means of a two-zone heating system attached to the outside of the SS shell. The annulus between the steel and tantalum vessels was filled with static helium (5 psig) to promote uniform heat transfer to the solution in the vessel. Each zone was heated by rod-type electrical cartridge heaters contained in a clamshell arrangement about the vessel. Thermocouples embedded in the clamshell between the individual cartridge heaters monitored heater temperature as well as temperature gradients within each heater assembly. The thermocouple readouts were used as input signals to the heater

controllers and also provided useful diagnostic information about the functionality and performance of the individual cartridge heaters. The clamshell assembly was designed to allow remote replacement of each heater or thermocouple. A separate kilowatt-hour power meter that measured heat input was included in the heater-control circuit for each dissolver. Input information was compared with heat-output data generated from the condenser cooling system as part of heat-balance calculations that were carried out to monitor overall dissolver performance. The lower and upper zone heaters on each dissolver could be operated separately so that, for example, only the lower zone would be heated during a low-volume (e.g., 1 L) dissolution. A blower was mounted adjacent to each dissolver vessel to cool the vessel with cell air at the end of a dissolution.

The dissolver vessel was closed by a gasketed flange assembly in which a Teflon-coated gasket was compressed between a tantalum flange welded to the top of the primary vessel and a tantalum plate attached to the bottom of a SS spider flange, which was connected by four swing bolts to a mating SS flange welded to the SS safety shell. This arrangement ensured that all wetted parts of the vessel consist either of tantalum or Teflon. The swing bolts were tightened with an air-powered impact wrench modified for remote operation (the wrench was counterweighted to facilitate transport with manipulators and is operated from an out-of-cell foot switch). Force applied to the bolts was controlled by adjusting, from outside the cell, the air supply pressure of this wrench.

A 32-in.-long tantalum rod was seal-welded to the bottom of the tantalum plate attached to the spider flange. Attached to this rod was a tantalum basket, which held the fuel-rod segment during its dissolution. The lower portion of the basket could be disengaged from the rod, by means of a bayonet-type mounting closure, to insert the sample can holding a given segment. In loading a segment, the entire spider-flange assembly was hoisted above the dissolver vessel with the in-cell bridge crane. After the segment was inserted into the basket and the basket was rejoined to the tantalum extension rod, the assembly was lowered into place and the closure seal made.

Reagents, solutions, and gases were transferred into and out of the sealed dissolver vessel by means of ports bored through the tantalum flange welded to the primary vessel. One of these ports was connected to a tantalum spray ring located near the top of the vessel; the spray ring rinsed down the vessel walls and the basket with fresh reagent. A second port was connected to a tantalum-tubing drain line (1/4-in. OD) that extended (internally) to the bottom of the vessel. Reagents and solutions could be removed from or added to the vessel through this line. The bottom end of the drainline had small rectangular notches cut in the tubing periphery. In the original vessel design, the drain line rested against the bottom of the vessel, and the notches served as a kind of coarse filter to prevent fuel particles and cladding fines that escaped the basket from leaving the vessel. In early tests of the system, however, some particles and fines passed through the notches while the vessel was being drained, and then clogged the drain line when reagents were added through the drain in the opposite direction. To overcome this problem, the end of the drain line was raised above the vessel bottom by about 1/8 in. (Bending of the drain line to raise it was done with the dissolvers in place in the hot cell. Specially built fixtures and jigs were used to ensure a minimum of hands-on effort; gamma activity in the cell at the time of these operations was low enough for suited entry into the cell.)

Another port in the tantalum flange led to the dissolver off-gas system, which controlled pressure for the system and collected gases released during the dissolution. These gases were subsequently mixed and sampled, and eventually mixed with cell exhaust air for slow release. Pressure control was provided by a dome-type valve (Grove Mity Mite, Model S-91LW; Grove Valve and Regulator Co., Oakland, CA) with a Teflon membrane.

A demister, in the form of a 500-cm³ SS expansion volume was inserted in the piping between the dissolver vessel and dome valve; its purpose was to remove condensed or entrained liquid droplets from the dissolver gases. The piping from dissolver to demister was pitched to permit liquid to flow back into the dissolver by gravity.

The outlet of the dome valve was connected to a nominally 60-L SS cylinder (the "off-gas hold tank"), which was fitted at its bottom with a valved drain line to remove liquids that might accumulate in the tank. Electronic pressure transducers and mechanical pressure gauges (for backup) were installed at various locations in the off-gas-system piping to monitor the dissolver pressure, hold-tank pressure, and pressure at several intermediate points in the piping. Thermocouples provided temperature data at several of these locations. An SS dual-bellows-type pump aided in transferring gases from the dissolver to the hold tank at the end of a dissolution. The same pump, through valving, was incorporated into a closed-loop gas recirculation system that allowed the gases collected in the hold tank to be mixed prior to sampling; the recirculation loop carried the gases through two 150-cm³ SS gas-sampling cylinders, which were connected in series and located in a hood (the gas-sampling station) on the outside of the DDS cell.

Each sample cylinder was fitted with plug-type closure valves and self-sealing quick-disconnect fittings for attachment to the off-gas system piping. Gas flow through the recirculation loop was monitored by means of an electronic flow transducer located in the in-cell piping.

The dissolver drain line was connected to a solution transfer system consisting of a six-valve manifold, an in-line solution filter assembly, and associated piping. This line transferred reagents and solutions into and out of the dissolver vessel. The valves were piped such that reagent from the reagent-charging system (see below) could be added to the vessel through either the spray ring or the drain line, while the in-line filter was back-flushed. Solution removed from the vessel through the drain line and filter assembly could be directed to the BT or to a sampling outlet. The valve manifold and the solution filter for each dissolver were located over an SS drip pan in full view of the work station window. This arrangement allowed for convenient replacement of the filter, as well as ready containment and visual inspection in the event of leaks developing in the solution transfer system.

The in-line solution filter consisted of a 10- μ m Millipore membrane in combination with a penetration-type prefilter mounted in a reusable SS housing. The entire filter housing was replaced as a pretested unit each time the filter was changed (a new filter was used for each segment dissolution); 19 interchangeable filter housings were cycled repeatedly during the POB operations. The filter housing was a commercial unit but was modified by machining off all internal corners and installing a fine-mesh screen in the inlet line. These

modifications improved the transport of fuel particles from the filter back to the dissolver vessel during backflushing operations with the system. The prefilter alleviated loading up of the membrane filter and consequent slow transfer of solution. A fixture for the remote disassembly and rebuilding of the filter housings was installed at the general-purpose work station in the DDS cell. This fixture included a vise and fixed-position pipe wrench for manipulating the threaded-collar closure of the housings and a gas manifold with pressure gauge for pressure testing the units after reassembly.

The BT into which solution and rinses from the dissolver vessel were transferred was one of nine 9-L SS vessels with dome-shaped tops. Each vessel was reusable and had a large-bore (0.506-in. orifice) gate valve for access into the vessel and appropriate fittings to mate the solution transfer system to an integral BT inlet tube. Each BT also contained an integral stirrer assembly that could be connected through a chuck coupling to a variable-speed stirrer motor mounted on a yardarm attached to the cell wall.

The BT rested in a stanchion-supported, insulated, bucket-like secondary vessel that rested in a SS drip pan on a recessed, top-loading, remotely operated, electronic balance. The secondary container featured a bottom-mounted ring heater (700 W) in direct contact with the BT and an air-line connection that directed cool air around the BT; these features ensured rapid heating and cooling of the BT solution. The stanchion support of the secondary vessel included four air-powered pistons that allowed the entire assembly to be remotely lifted off the balance, either by a manual switch or by computer command, so that the balance could be tared or repetitive weighings of the assembly could be conveniently performed. The stanchion mounting also allowed manual or remote tipping of the BT to one side, which facilitated removal (by use of a portable drain tube inserted through the gate valve) of all but a few milliliters of solution from the tank. The drip pan included in the BT assembly protected the BT balance from spilled solutions, dropped tools, etc. The balance was a Mettler Model PK-60 having a capacity of 60 kg and readability of 1 g.

b. Reagent Charging System

Reagents used with the DDS were prepared in 50-L batches by the ACL and were stored in 50-L, screw-cap, polyethylene bottles. They were introduced into the dissolvers by means of the reagent-charging subsystem. This subsystem consisted of a reagent-dispensing station located in a fume hood adjacent to the DDS hot cell, a distribution pump, and SS piping that carried the reagent to the solution transfer system of either of the two dissolver systems.

The reagent-dispensing station comprised (1) a reagent selector switch, which sets appropriate valving to connect a peristaltic metering pump to one of three reagent storage bottles located in a deep SS catch pan under the hood, (2) pump controls, and (3) a charging vessel mounted on a top-loading electronic balance (Mettler Model PS 30; 30-kg capacity, 1-g readability). Reagent from the selected storage bottle was pumped into the charging vessel until a desired amount was obtained, as indicated by the balance readout. The reagent was then pumped by the distribution pump into the appropriate dissolver after the in-cell valving had been set to charge the reagent through the appropriate flow path. A pressure gauge in the reagent distribution line monitored the line

pressure (typically 30 to 40 psig) during reagent addition. Reagents other than the three usual dissolver reagents (e.g., distilled water) could be charged to the cell by dispensing them directly into the charge vessel.

c. Vacuum-Assisted Solution Transfer System

Solution was transferred from the individual dissolver vessels to their respective BTs by evacuating the BT and venting the dissolver vessel. The differential pressure moved the solution. The vacuum source consisted of a 25-L vacuum reservoir tank and an SS, single-bellows-type (vacuum) pump mounted downstream of the tank. The reservoir was evacuated and isolated from the pump before being opened to the BT assembly; this avoided damage to the pump from corrosive solution vapors and provided a closed system in which dissolver gases that might escape the solution were contained. Each dissolver system had its own vacuum-reservoir system.

d. BT Solution Sampling System

Samples of solution from the dissolver BT were required for analyzing the dissolved segment fuel. The system used for withdrawing these samples from the BT is illustrated in Fig. 5. It included a variable-speed, reversible, peristaltic pump; a disposable sampling assembly (described below); and an air-actuated bottle-clamping fixture to hold the sample bottles. The system was designed to minimize solution losses during sampling, to be conveniently operated remotely, to provide approximately the same volume of solution in each bottle, and to avoid sample contamination.

Fig. 5. Blend Tank Solution Sampling System

The sampling assembly consisted of a rubber stopper, sized to fit the bottle opening, through which two sections of 1/8-in. OD polyethylene tubing were inserted; a new sampling assembly was used each time the BT was

sampled. One tubing section was long enough to be inserted into the BT through the wide-bore gate valve on the tank; the other end of this tubing protruded through the stopper by a length equal to the height of the void space in a properly filled bottle (about 25 mL, or 35 g, of solution was taken for each sample). The other section of tubing extended only to the bottom of the rubber stopper, and connected to the peristaltic pump. The pump moved only air into or out of the sample bottle. Solution from the BT was drawn into the bottle by pumping air out of the bottle; when the protruding end of the inlet tube was submerged in solution, the pump was reversed, and air pressure in the bottle returned excess solution to the BT, clearing the inlet tube of liquid. The stopper/tubing assembly could then be moved to another bottle and the process repeated.

A major advantage of not pumping the solution directly was that tubing in the pump heads did not have to be replaced to provide clean sampling lines; frequent remote assembly and disassembly of the peristaltic pump heads would have been an onerous task. The stopper/tubing assemblies used on this system were prepared en masse outside the hot cell and transferred in as needed.

The bottles used to contain the solution samples are also worthy of note. We selected round, 60-mL Pyrex bottles with a narrow-mouth screw cap closure. Polypropylene caps were used on the bottles after early testing showed that Bakelite caps changed weight with changes in ambient conditions, possibly because wood flour is used as a filler in their manufacture. The polypropylene caps were, in addition, modified to permit venting of radiolytically produced gases generated in the solutions. This modification was made to avoid pressure buildup in samples placed in archive storage. To provide the venting, a small (1/16-in. dia) hole was drilled in each cap and a liner, consisting of a gas-permeable membrane and a Viton sealing gasket, was inserted against the inside of the cap.

With 25 mL of solution, a bottle could be laid on its side without having solution enter the bottle neck; liquids did not easily penetrate the gas-permeable membrane, even with the bottle held upside down. Of course, the solution samples were not intentionally handled in such ways, but selections were made to accommodate worst-case situations.

e. Liquid-Waste Storage System

Provision was made for storing, in the DDS hot cell, the fuel-bearing waste solution produced by the dissolvers. The liquid-waste storage system centered around two 370-L SS waste tanks located between the two dissolver systems. Each tank rested in a deep secondary tray and was enclosed with lead cover plates (1.0-in. thick) to shield the in-cell equipment and thereby minimize damage to radiation-sensitive items. Each tank was also equipped with an externally controlled mixing system and instrumentation to measure liquid level. The vapor space of each tank was continuously purged with house-supplied air to enhance the evaporation of stored solution and thereby reduce the amount of liquid waste needing to be processed into cement waste. A vent delay was incorporated into the air outlet to allow for the holdup and decay of ^{220}Rn (a daughter of ^{232}U), which emanated from the waste solution.

The two waste tanks were connected to a system of valves, which allowed a common transfer pump to be used in moving solution from either BT

to either waste tank, from one waste tank to another, or from either waste tank to the waste treatment facility located on the floor above.

f. Weighing Systems in the DDS Facility

The DDS facility contained five remotely operated electronic balances. Two of these were included as part of the BT assemblies as described in Section III.A.4.a; each BT balance had a set of three 5-kg standard weights fabricated at ANL (see Section III.A.2.d for fabrication details). The other three balances were located in balance wells in the cell work table as described in Section III.A.2.d. These included two PK-300 balances (one at each dissolver work station), which were used to weigh segments (<286 g) and solution samples. The remaining balance was a PK-2000 unit used for weighing segments of mass greater than 286 g prior to their dissolution; this balance was located near the general-purpose work station of the DDS cell.

5. The Analytical Hot Cell

The Analytical Hot Cell was a kilocurie cell having two work stations and was equipped for preparing samples, generated in the POB operations, for analysis. These preparations included such operations as dividing solution samples into weighed aliquots, performing solvent extraction and ion-exchange separations, and carrying out a variety of chemical transformations on samples and solutions. The cell contained two top-loading electronic balances: one, used for coarse weighings, was a Mettler Model PR-700 (dual range, either 70-g capacity at 0.01-g resolution or 700-g capacity at 0.1-g resolution); the other was a remotely operated Model AK-160 (160-g capacity at 0.1-mg resolution), which was located in a balance well with draft shield and interfaced to the project data system. The AK-160 balance was used for weighing the samples and aliquots. Two large-surface hot plates, a clinical centrifuge, and a variety of reagent dispensers and sample-handling fixtures also were in the cell.

In addition to the transfer systems installed in the alpha barrier (see Section III.A.2.b), this cell also had a double-door sliding transfer hatch built into one of its shielding walls. The transfer hatch was a 10 by 12 in. opening in the cell wall, with pneumatically powered shielding doors covering the in-cell and out-of-cell faces of the opening. The door controls were interlocked so that only one of the two shielding doors could be open at any time. To provide easy access to transferred items, the opening had a sliding tray that could be extended into or out of the cell (when the appropriate door was open). A Lexan and steel transfer box, equipped with two glove ports and a SS bottom tray, was attached to the outer cell wall over the shielded transfer hatch opening. The transfer box was vented into the cell and served as an interface in transferring miscellaneous small items such as beakers, ion-exchange columns, etc., into the cell. In addition, it was used for moving prepared samples out of the cell to other analytical facilities. A shielded area for temporarily storing prepared samples (e.g., samples for gamma-counting analysis) was constructed from lead bricks on the floor of the transfer box.

6. The Waste Treatment Facility

A "hot cement" works was set up to dispose of analytic residues (mainly, the bulk dissolver solution) as a monolithic TRU waste form. The cell directly above the DDS cell was selected for the waste treatment operation to

simplify transfer-piping installation. The cell was partitioned so that it formed an L-shaped alpha-containment region, which was further divided to separate each leg of the "L." Each cell area served a different function. The larger area housed two specially designed mixing stations, a transfer lock for moving materials into the cell, and a specially designed "can-out system" for removal of cement-filled cans, which comprised the monolithic waste. The smaller area contained 10 drying ovens (see process description below). The barrier wall separating these two regions was fitted with a transfer lock but also allowed for directed ventilation flow (also discussed below). All of the equipment was designed to be maintained remotely with M/S manipulators. Because waste treatment operations did not affect the validity of ANL's destructive assay results, discussion of this topic is kept brief in this report. Consequently, facilities, operations, and results are all described in this section (III.A.6), rather than being separately considered as is done for the other major operations areas (FSS, DDS, Analytical).

a. Equipment

(1) The Cement Mixing Stations

The two cement mixing stations were designed and built at ANL. Basically, they were Unistrut-framed, Lexan plastic boxes, each fitted with a relatively tight-fitting, gasketed door. Box features included a specially designed paddle for efficient mixing in the paint-can type primary containers; the paddle was driven by a gear transmission drive. A fixed, water-cooled reflux condenser was mated to a portable spool section that was brought in with the primary can; this minimized acid vapor release to the cell ventilation system (and cell stack). A pneumatically driven sliding/lifting platform raised the primary can into place for mixing.

(2) Auxiliary Equipment

Auxiliary equipment included an acid-feed system for slowly adding waste solution to the cement during mixing and 10 drying ovens for dehydrating the cement product. The feed system was built around two 11-L acid-feed reservoirs, each fitted with liquid level probes and an adjustable overflow line so that the liquid inventory could be limited to the desired quantity, nominally about 2.5 to 3.0 L; excess liquid was returned directly to the waste storage tanks in the DDS cell. Peristaltic pumps transferred the acid waste from the DDS cell up to the feed tanks in the cement-making cell and metered the solution sent to the cement mixing station. Two simple beam balances monitored water removal in the course of the setting and drying steps.

The drying ovens (product of Sybron/Thermolyne, Dubuque, IA, Model No. 0V19225) operated from 120-V AC line voltage with a power rating of 350 W. Each oven was modified to facilitate remote operation and repair. The door latch was replaced with one that could be more easily operated with M/S manipulators; the heating element was mounted in a drawer that could be easily slid into or out of the oven cabinet while the element connectors were being aligned; and the oven thermostat was removed, temperature control being provided by an out-of-cell controller connected to a thermocouple that was installed in the oven.

b. The Waste-Cement-Making Process

The process for making the cemented waste evolved after a review of literature, evaluation of waste disposal options, and laboratory formulation studies. This process is considered unique in at least two ways: (1) thin-walled steel cans were used for the disposal of a 10-N acidic ($\text{HNO}_3\text{-HF}$) solution; and (2) the filled cans, after setting, were dried (at 125°C) to reduce the residual water content so that pressurization due to radiolysis effects would not present a hazard.

The several-step process is outlined below.

- (1) A weighed cement-water-lime slurry containing small amounts of two commercial additives, an anti-slumping agent, and a set retardant is premixed in a nominally 1.5-gal steel can (similar to a paint can).
- (2) The can (with lid) is transferred into the mixing area of the cell by way of the transfer hatch. A specially designed can lifter is used with the manipulators because the cans have no bails (a space-saving feature for the packaging step).
- (3) A spool piece for connecting to the condenser in the mixing station is placed on the can, and the can is placed in the mixing station and raised into position for the acid-addition mixing step.
- (4) A 2.8-L batch of waste solution is transferred up from the waste storage tank in the DDS cell to the appropriate hold tank in the cement-making cell.
- (5) The mixer is turned on, and the acid feed started. (The duration of the feeding period was reduced from three to two hours during the course of the waste work-off. A magnetically held surface thermometer on the side of the can monitors the temperature of the mix; temperatures to 90°C were observed.)
- (6) After the mixing step, the can is weighed and set aside for the contents to set, nominally for three days.
- (7) The can is then weighed again and oven-dried (at 125°C) for four days.
- (8) Several cans are accumulated, and subsequently topped-off with additional hot mix to increase the overall waste loading per can. (Loading is increased $\sim 25\%$ by this step.)
- (9) The topped-off cans are again allowed to set and then given a final drying for up to seven days, or until a constant weight is achieved.

Interlocks and other safety features were incorporated to ensure safety in carrying out the process, as follows:

- (1) The platform could not be raised unless the door to the mixing box was fully closed and latched.
- (2) A mechanically set restraining pin was used to lock the platform in the "up" position; thus, in the event of air failure, the platform would not slide down while mixing was in progress.
- (3) The acid feed pump and holding tank outlet valve were automatically shut off if the mixing motor stopped. An audible alarm was included in this system.
- (4) A float-type liquid sensor in the drip pan under the acid-feed pump also halted the acid feed and shut off the pump in the event of liquid buildup due to leakage.

c. Operations

Actual cement-making operations were started on November 11, 1984, and continued through October 1986. Some 700 L of waste dissolver solution, producing ~230 cans of waste, was disposed of. In addition, the cement approach was used to dispose of non-transuranic (TRU) wastes, e.g., Zircaloy hull residues from the dissolver operations (mixed with cement in a 1:1 weight ratio), and a basic aluminum nitrate solution from the analytical cell. For the non-TRU wastes, the mixing was done by hand (i.e., with the manipulator and a paddle) rather than in the mixing station.

Operations were generally smooth, except for one recurring problem, namely, plastic tubing failure, either in the peristaltic pumps or at end-fittings. The tubing apparently degraded due to the severe conditions, i.e., radioactivity plus strong acid. Its failure resulted in leakage, generally requiring localized cleanup (tissue mop-ups), which created another small waste stream. In time, a maintenance schedule was established to periodically change the pump line, but even this precaution did not prevent failures occurring from time to time.

Simple and effective means for disposing of three side waste streams were developed during the course of the cement-making operations. Disposal of tissue used in wiping up solution spills was first attempted by forcing the bulky wads of paper into wet cement in the primary waste cans. This proved quite awkward when working with manipulators. The alternative route explored was thermal degradation. Bench tests showed that tissues degraded essentially to a powder at about 140°C. Since our drying ovens had a limit of about that level, it was a simple task to loosely wad dried tissue into an empty waste can in the cell, heat the material for a few hours, and then use this can, which contained the degradation product and a small amount of associated nitrate salts, to prepare a normal can of waste. This method proved successful.

Cans used in preparing the cement with which other cans were topped off accumulated in the cell and represented the second side waste stream. To dispose of these cans, we began preparing the starting cement slurry in-cell

with these cans. The dry slurry components were brought into the cell in plastic pouches, and a water supply for forming the slurry was provided. After some preliminary "hand" mixing of the slurry in a given can, the can was installed in one of the mixing stations where it was treated as described earlier. All cans that had accumulated inside the cell during early waste disposal work were disposed of in this manner.

A third side waste-stream was the plastic pouches used in transferring the slurry components and other polyvinyl-chloride-plastic waste material. Again, the drying ovens served a useful purpose because the plastic had a relatively low melting point. The plastic was loaded into an empty can and the contents were heated for about two hours, giving a consolidated, rather high-density mass (much like a cake of wax), readily disposed of as a radioactive, but non-TRU waste. No problems were encountered with off-gasses from the plastic.

d. Packaging the Waste

To facilitate disposal of the waste at an away-from-ANL repository, a waste package ("package" refers to the item placed in actual storage at the repository) was designed to meet requirements for TRU waste storage at the federal Waste Isolation Pilot Plant (WIPP). The requirements include use of two alpha seals, limited (to 7 psig) pressurization potential, and use of approved containers. Further, for contact-handled waste, radiation levels at the surface of the package had to read <200 mR/h. Because our primary cans of waste gave readings of ~ 250 R/h, a shielding cask was necessary (see below).

The package included the following:

- (1) Two sealed primary cans loaded vertically into a secondary can (similar to a paint can). The lid on the secondary can serves as one of the two alpha seals.
- (2) A steel shielding cask that provides a snug-fitting cavity for the loaded secondary can and has about 2.5 in. of lead shielding. The lid on the cask is bolted but not gasketed.
- (3) The outer container, an approved (DOT-17C) galvanized 55-gal drum.

Shipment to an interim storage site was necessary because the WIPP has not been completed; the drums were loaded into Polypanther (trade-mark) overpacks, six to a truck, and are being sent to Rockwell-Hanford, which can store the waste for up to 25 years.

B. Analytical Facilities

Facilities associated with the ACL and used in the POB project included four laboratories equipped for general or specialized analytical chemistry operations and an alpha/gamma counting station installed at the Analytical Hot Cell specifically for use with project samples. The laboratory facilities included (1) a general-purpose laboratory for preparing chemical standards and reagents needed in project operations, (2) a Uranium Purification Laboratory set up for isolating and

purifying alpha-active samples for mass-spectrometric uranium analyses, (3) the ACL Thermal Ionization Mass Spectrometry Laboratory, wherein uranium isotopic measurements were made, and (4) the ACL Gas Mass Spectrometry Laboratory, in which the composition and fission-gas isotopy of POB gas samples were determined.

Key equipment items contained in these facilities as they relate to project measurements and operations are described in the following subsections.

1. General-Purpose Laboratory

The laboratory used for preparing reagents and chemical standards for the POB project was a fully serviced chemical analysis laboratory that contained several fume hoods for handling radioactive or corrosive materials. The laboratory was equipped with three Mettler balances: (1) a Model H-315 mechanical balance (1-kg capacity, 0.1-mg readability); (2) a Model HK-60 hanging-pan electronic balance (30-g capacity, 0.01-mg readability); and (3) a Model AK-160 top-loading electronic balance (160-g capacity, 0.1-mg readability). The two electronic balances were interfaced to the project computer system, as was a video display terminal located in the laboratory.

2. Uranium Purification Laboratory

A laboratory near the Shielded Cell Facility was set up to perform separation and purification chemistry on uranium samples, which were previously separated from gamma-emitting sample components in the Analytical Hot Cell. This laboratory was equipped with a full complement of typical laboratory services but also contained a five-unit train of hoods approved for handling alpha-emitting nuclides.

The hood train had small equipment items such as vortex mixers, a centrifuge, support stands for ion-exchange columns, hot plates, and a Meeker burner. One of the hoods also contained an apparatus (Kepco Model CC-15-1.5 M) for the constant-current electrodeposition of uranium on SS planchets, which was used for preparing portions of each uranium isotopic sample for alpha pulse height analysis. This apparatus had two independent current supplies so that two cells could be run simultaneously.

Cells for the electrodeposition were fabricated at ANL from 1-oz. polyethylene, screwcap bottles machined to accept electrode assemblies composed of Teflon supports and platinum electrodes. In fabrication of each cell, the bottom of the bottle was cut out to accept a Teflon plug that positioned a platinum counter electrode; a 3/8-in. hole was drilled in the bottle cap to accommodate a gasketed planchet holder, which sealed the planchet against the bottle's neck when the bottle and cap were screwed together. Because only the bottle and the planchet were exposed to solution during the electrodeposition with this cell design, the cap assembly could be reused repeatedly without a threat of sample cross-contamination. A large supply of machined bottles was maintained to permit the use of a new cell body for each sample processed.

3. Thermal Ionization Mass Spectrometry Laboratory

The Thermal Ionization Mass Spectrometry (TIMS) Laboratory features two mass spectrometers, both having magnetic-sector mass analyzers and deep Faraday cup detectors. One of these instruments, an Isomass 54R manufactured by V. G. Isotopes, was procured and set up specifically to meet the rapid pace and demanding requirements of the POB analytical program. This instrument is fully automated and capable of sequentially analyzing up to 16 samples without operator intervention once the measurement sequence is initiated. It features fixed source and collector slits; a 16-position, rotating-turret source pumped with a 270 L/s turbomolecular pump; and a digital integrator readout circuit. It is not equipped with a filament preheat supply. The liquid-nitrogen cold trap on the ion source was modified to increase its capacity and thereby permit overnight operation without attention to the trap. Operation of the instrument is controlled by a Hewlett-Packard (HP) 9845-B desktop computer having 187 kilobytes of memory, two integral tape drives, and a floppy disk. The HP 9845-B computer was interfaced to the project computer system as described in Section III.C.

The other instrument in the TIMS laboratory was manufactured by Teledyne Isotopes according to NBS design.^{6,7} It is a manually operated instrument connected to a HP-9820 desktop computer for data acquisition and analysis. It served as a backup for the VG 54R instrument, and was also used routinely for the analysis of POB samples that required unique operating conditions or special operator attention during a run.

To support the operation of these instruments, the TIMS laboratory also contains special areas for storing samples and loading the filaments on which samples are introduced into the instruments. To minimize chance for cross-contamination, samples are stored in a fume hood separate from the work stations in which filaments are loaded and mounted in mass spectrometer source assemblies. For the POB uranium samples, an area within this hood was set aside and surrounded with lead bricks to shield against gamma rays emitted by daughters of ²³²U present in the samples. The work stations for loading and mounting the filaments are set up in two exhausting, Class 100, clean air hoods (Environmental Air Control, Model LAB-1). One work station is equipped with a programmable sample dryer, fabricated by ANL's electronics shop, which follows a reproducible sequence of heating steps (by passing preset currents through the filament) to dry and properly oxidize the uranium on each loaded filament.

4. Gas Mass Spectrometry Laboratory

The chemical and isotopic composition of gaseous POB samples was routinely determined in the ACL's Gas Mass Spectrometry Laboratory by one or both of the two different instruments in this facility. The sample inlet section on each instrument had an adapter that permitted (1) direct attachment to the quick-disconnect fittings on the POB gas-sampling cylinders and (2) sequential expansion of the gases to reduce their pressure to a value compatible with instrument requirements.

The instrument most used for POB gas measurements was a cycloidal mass analyzer, Consolidated Engineering Corp. (CEC) Model 21-620, having a modified electronics package, an Esterline-Angus E11015 strip chart recorder,

and a CEC micromanometer, Type 23-105, for monitoring source pressure. This instrument exhibits good resolution and sensitivity and permits relatively convenient determinations of chemical composition.

The other instrument used for POB samples was a Nuclide Analysis Associates Model 6-60, RMS-7, also having a modified electronics package and an identical strip chart recorder. The Nuclide instrument is designed for isotope-ratio measurements and features a magnetic-sector mass analyzer and dual (plate-and-cup) detector system. Source pressure is monitored with an MKS Instruments, Model 170M, micromanometer. This instrument has better resolution and somewhat higher sensitivity than the CEC 21-620 and permits more-reliable determination of the isotopic composition of an individual component in a mixture. When operated in a scanning mode with the cup detector, it also provides quantitative information on the sample composition and may be used to corroborate measurements performed with the CEC 21-620 system.

5. Alpha/Gamma Counting Station

A fully automated alpha/gamma spectrometer system was installed in a work area adjacent to the Analytical Hot Cell. This system consisted of a microprocessor-controlled, 26-position, automatic sample changer (built at ANL), along with a series of components purchased from EG&G ORTEC. These other components included a Model GEM 10175 high-purity germanium (HPGe) gamma detector; a Model 576 dual-port alpha spectrometer featuring a separate, ruggedized-surface-barrier alpha detector in each port; and a Model 7050 Data Acquisition and Analysis System, which constitutes a sophisticated multichannel analyzer (MCA) and terminal control console for acquiring, displaying, and manipulating counting data. Our system included three separate analog-to-digital conversion (ADC) subunits, which simultaneously acquired the output from the gamma detector and each of the two alpha ports in separate segments of the MCA memory; 4096 channels were used for the gamma ray spectrum and 250 channels for each alpha spectrum.

The sample changer and detectors were located in a small isolation room, which also contained the dead-air transfer box through which prepared samples were removed from the Analytical Hot Cell. Placement of the gamma counter in this location minimized the distance over which the radioactive gamma-counting samples had to be transported.

The MCA and control console for the system were installed in a control room that was built of fiberglass and Plexiglas panels in the bay area at the face of the cell. This room also contained the PDP-11/23 computer (see Section III.C) to which the alpha/gamma spectrometer was interfaced.

C. The Project Computer System

Two minicomputers and a desktop computer (dedicated to the project), plus a super-minicomputer (shared with several hundred other users), were used for process control and monitoring, data acquisition, and data reduction and management. The minicomputers and the desktop computer were interfaced directly to process and measuring equipment within the project, including the FSS, DDS, TIMS, and alpha/gamma spectrometer. The super-minicomputer was not involved in laboratory operations but was used to receive, collate, and store batches of data from the three smaller computers and to generate reports.

Each operations area within the project was equipped with a computer work station, which included video display terminals (VDTs) and printers as appropriate for the operations performed.

Software used with the systems included that supplied by the hardware vendors, as well as many programs with general and specific applications developed at ANL for the POB project.

Discussed below are the hardware that constitutes the project computer system and the general-purpose software. Specific applications programs are discussed in later sections that treat operations in which the individual programs were used.

1. Hardware*

A schematic representation of the project computer system is shown in Fig. 6. The hub of the system is the VAX-11/780 central processing unit (CPU) located in the Chemical Technology (CMT) Divisional Computing Facility. This computer system has approximately 2000 megabytes of disk storage, two tape drives, and a variety of high-quality peripherals for reporting data, such as a

Fig. 6. Computer System for POB Project

*All computer-related devices are products of Digital Equipment Corp., Maynard, MA, unless otherwise noted.

high-resolution pen plotter for graphics and a laser printer for letter-quality text output. The VAX-11/780 is connected to the two project minicomputers through dedicated telephone lines and DECNET software. The desktop computer (i.e., the HP-9845-B system attached to the VG 54R mass spectrometer, Section III.B.3) is connected to the VAX system according to IBM's 2780/3780 protocol, which is a de facto industry standard and is supported by both manufacturers (i.e., HP and DEC).

One of the minicomputers (designated "POB") is located in a control room that was constructed in the bay area adjacent to the DDS hot cell. This system features a PDP-11/73 CPU and three RL02 disk drives and is interfaced to the DDS, the FSS, and associated balances by a CAMAC (Computer Automated Measurement and Control, an IEEE standard) bus. Serial interfaces to all project balances were adopted for ease of connection (these require fewer conductors in comparison to parallel interfaces) and programming. The control room also contained an LA-120 printer, set up as a console device for the PDP-11/73 minicomputer, and a VK100 (GIGI) terminal for general use. This terminal was connected to a Gandalf PACX-IV terminal switch associated with the VAX-11/780 system and could thereby access the VAX system or either of the two minicomputers.

The second minicomputer (designated "GAMMA") was located in the control room adjacent to the Analytical Hot Cell along with the control console of the alpha/gamma spectrometer (see Section III.B.5). This PDP-11/23 minicomputer included two RL02 disk drives and was connected to an LA-34 printer for local hardcopy output. It was interfaced to (1) the alpha/gamma spectrometer, (2) the automatic sample changer on the gamma-counting system, and (3) three electronic balances, namely, the remotely operated AK-160 in the analytical hot cell and the two balances in the ACL General-Purpose Laboratory (Section III.A.1); for these latter balances, which were located in a different wing of the building, dedicated telephone lines were used for the connection.

Each of the major operations areas within the project was equipped with a complement of terminals and printers to provide communication with the computer system and to generate hardcopy operations records as appropriate. The FSSF work station included a VT-125 text-and-graphics terminal plus a LA-50 printer, chosen for its small size, compatibility, and reliability. Identical printers were provided for each dissolver system of the DDS (DS1 and DS2); each dissolver work station was further equipped with two terminals, one VT-102 text terminal, and one GIGI color-graphics unit.

Terminals at the analytical hot cell included a VT-102 text terminal used for operations at the cell work stations and the console terminal of the alpha/gamma spectrometer. This console terminal, a Tektronix Model 650 (product of Tektronix, Inc., Beaverton, OR) graphics monitor, was normally used for display of spectra but also served as a text terminal for the GAMMA computer.

Besides being accessible from the terminals at the various project work stations, the two minicomputers could also be reached by password access from office terminals connected to a PACX-IV switching system. This feature provided managerial access to project data for review and monitoring purposes; however, software interlocks built into the system prevented operation of process

equipment from any terminal other than that located at the work station by which the equipment was served.

2. Software

Software incorporated into the project computer system included software systems (i.e., collections of interrelated programs) supplied by the hardware vendors for use with their equipment, as well as systems and programs developed at ANL for the project. Some of the vendor-supplied software was customized to meet the program interfacing needs of the project, to support the CAMAC and other hardware interfaces, and to provide data in a form consistent with project data management and reporting requirements.

Software used by the project falls into three categories: (1) operating and communications systems, (2) applications systems, and (3) stand-alone programs. The operating system manages allocation of processor, main memory, and disk- or tape-storage resources and provides the organizational structure for the computer file system; communications programs supply the means for transferring data across communications lines and interfaces. Applications software systems use the foundation provided by the computer operating system to carry out specific tasks involved in the acquisition, manipulation, and storage of data (e.g., operate drive equipment, read and write disk files). Stand-alone programs perform the same function but are restricted to executing a rigidly defined sequence of tasks in only one way. The interactive execution of interrelated programs within the applications system permits flexibility and generality to be achieved by allowing the sequence of tasks to vary according to information made available to the system as it is running.

Detailed consideration of each of the software items in the project computer system is far beyond the scope of this report. However, in the following subsections, we identify the major software systems and programs and briefly describe their purpose and structure. Additional information related to some of the programs may be found in the data-management and operations discussions presented later (Section IV, especially).

a. Operating and Communications Systems

Operating system software on the four computers associated with the project was different for each type of computer. The VAX-11/780 system is based on a 32-bit word size and runs DEC's VMS (virtual memory) operating system. Both PDP-11 computers (11/23 and 11/73) operated under control of the latest version of RSX-11M. The RSX-11M is a real-time multitasking operating system, which can rapidly respond to external events and is capable of running multiple programs simultaneously. Among the additions that were made to this system at ANL for the POB project was a program to speed up and direct data communications between programs operating on the system; this program (VS for Variable Send) significantly improved system performance and has been distributed (on request) to a large number of RSX-11M users around the country for incorporation into their systems.⁸ The operating system running on the HP-9845B desktop computer is a fixed system stored in read-only memory and features BASIC programming language enhanced by the manufacturer.

Transfer of files between the PDP-11 systems and the VAX-11/780 was accomplished by DECNET software, a network-communications package that allows DEC computers to transfer data over a variety of communications lines and interfaces. Communication between the HP-9845B and the VAX-11/780 was facilitated by use, on each system, of software packages that emulate the IBM 2780/3780 communications protocol.

b. Applications Software Systems

Applications software systems used by the project computers included a variety of DEC-compatible software packages such as word processors, text editors, graphics packages, etc., that were available within the CMT Divisional Computing Facility on the VAX-11/780. In addition, there were three major systems used in executing the project operations: (1) the HP-9845B software package provided by the vendor of the VG 54R TIMS instrument; (2) the software package provided by the vendor of the alpha/gamma spectrometer system; and (3) a general procedure-execution software system (named "the POB system" or "POBSYS"), written at ANL for implementing the shearing and dissolution operations. They are discussed individually below.

(1) Thermal Ionization Mass Spectrometer Software Package

A full complement of programs for the HP-9845 desktop computer was provided by V. G. Isotopes with the 54R mass spectrometer. These programs included diagnostic and operating programs, as well as support programs to establish data files needed by the primary programs in performing their tasks. Portions of the instrument operation programs were modified extensively at ANL, particularly those sections dealing with sample identification, data acquisition and reduction, and reporting of results. The changes were designed to aid the instrument operator in assigning specific analysis sequences to individual samples, to tailor data-taking and -reduction routines to the kinds of samples anticipated for POB measurements, and to output results in a manner consistent with project data-management requirements, e.g., obtaining a format compatible with subsequent computer manipulation or maintaining redundant records for data security.

The modified programs were set up to recognize and interpret the project sample identification format and to use the information in the sample designation for assigning a particular analysis sequence to each sample. This analysis sequence included selection of the appropriate reference mass in calculating isotope ratios. Special calculational algorithms were used if an internal standard was added to the sample [see Section IV.F.1.d.(1) for a description of the internal standard procedure]. The program formatted output files to which the data would be written. Copies of the output file were stored on disk and on tape, and a printed record (having more detailed information than the computer file) was produced for each analysis.

(2) Alpha/Gamma Spectrometer System Software Package

Software for the alpha/gamma spectrometer centered around the GeLiGAM analysis software package, which was supplied with the spectrometer system by the vendor, EG&G ORTEC. This package is designed for manipulating data from gamma-ray spectra obtained with germanium-based gamma

ray detectors. The system is modular in structure and has many user-selectable options such as peaked background subtraction and automatic decay corrections.

All of the data reduction routines of GeLiGAM operate on data files stored on disk. Provision is included to create a spectrum file from the counting data acquired by the MCA of the spectrometer system, so that the data reduction routines can operate on the spectrum information.

Supplemental programs were written at ANL and added to the GeLiGAM package to accommodate our use of an automatic sample changer on the gamma system, to convert the output format of the data reduction programs into formats compatible with the POB data-management system, and to analyze spectra obtained from the alpha-counting system, since the ORTEC programs did not provide for this analysis.

(3) The POB System (POBSYS)

The integrated software system called POBSYS was developed at ANL to provide computer control of the complicated, lengthy, and often-repeated FSS and DDS operations. It is a modular, general-purpose system designed to carry out conventional process control, data acquisition, and data management for a detailed step-by-step operating procedure. Specific functions required for the execution of a general procedure, e.g., sensor monitoring, process control, data management, etc., are isolated within separate, essentially generic programs in the system. Application-specific information is provided to these programs from information files that are generated and read by means of other system programs.

A schematic representation of POBSYS is given in Fig. 7, which shows the six major programs (large circles) and four types of files (boxes) that make up the basic system. The "slave" programs (small circles) are subroutines that execute a given step (or steps) in the operating procedure and are individually prepared to satisfy the specific requirements of a given application. In our system, approximately 300 slave routines were involved in running the FSS and DDS, although POBSYS is not limited in the number of slaves that it can handle. The Master Procedure Execution Program implements the execution of the procedure by stepping through the procedure file, calling up slave programs appropriate to each step, and directing the other system programs to perform actions called for in the step. These actions might include acquiring data through the sensor monitoring program, changing setpoints for alarms or other sensor-monitoring criteria contained in the sensor-monitoring-criteria file, modifying text or graphics displays on the computer terminals, or retrieving data that were logged in a previous step and needed in a current calculation or comparison.

The contents of the files and slave programs are tailored to meet the specific requirements of a given application by means of a number of software "tools"—programs used to set up the process-control system but not usually executed during actual production work. These programs are not shown in the figure. They include the following: programs that translate operating procedures between the human-readable (text) format and the machine-readable (binary) format; a program that shows the current values of all sensors in a

Fig. 7. Block Diagram of POBSYS Software System

dynamically, updated display; a program that allows sensor table entries to be displayed and edited; programs that format and print selected entries from the binary-formatted operations logs created and maintained by the Master Procedure Execution Program; and programs specific to the RSX-11M operating system that implement the overlay scheme for slave routine execution, whereby one slave routine in memory is replaced by another.

Several features incorporated into the design and utilization of POBSYS proved highly advantageous during implementation of the computerized procedures for the FSS and DDS. Possibly the most valuable of these was the modular structure of POBSYS, which permitted the development of the shearing and dissolution procedures in parallel, each being constructed from individual steps defined by a specific step order, work-station display format, slave routines, sensor-monitoring requirements, and optional looping or branching information. The flexibility afforded by POBSYS allowed work on each procedure by several different programmers with a minimum of duplicated effort. In addition, individual portions of each procedure could be modified separately from other portions with no need for extensive recoding or recompilation of the software. These features greatly facilitated the making of changes to the computer software as each procedure was developed.

Another POBSYS characteristic that aided in developing the computerized operating procedures was a "read-only" test mode, which provides process simulation without affecting installed hardware. Test code within POBSYS can be enabled for diagnosing problems or for testing. The test modes include "read-only execution," in which process-control operations are inhibited such that software commands affecting hardware conditions are ignored, and "sensor simulation," which allows the test operator to specify simulated values for sensor readings. These features permitted development and testing of the process-control software to proceed without putting the process hardware in jeopardy. In some cases, the software was tested before the hardware system was completely built and available. During actual operations, the system could be tested off-line for troubleshooting purposes.

Other POBSYS features contributed to effectively managing the FSS and DSS operations and to ensuring the acquisition and preservation of quality data records. Operational control within POBSYS recognizes a multilevel hierarchy, with specific capabilities or privileges available at each level. Thus, ordinary operators may be allowed to execute and "sign-off" procedure steps under normal conditions, but actions that bypass normal status checks or override procedural restrictions may require a supervisor's authority. Similarly, programmers were not allowed to perform procedure steps unless operating in test or read-only mode. The POBSYS operates under the concept of documented responsibility--each action, such as completion of a procedural step, requires a signoff that records the time and person responsible for the action.

All data recording and manipulation operations are coordinated by a data management program specially designed for POBSYS. This program has the following features: sharing of file access among several system components; locking of individual records in a file for update operations; automatic hardcopy logging of data sent to files regarded as "critical" (to provide a backup in case of system faults and to give the operator an accessible reference copy of critical data); and quick retrieval of recorded data by using binary search techniques on ordinary sequential files. Thus, the data management program facilitates information flow between POBSYS programs and the files it manipulates, and provides redundant recording for data security. Also, the operator can enter comments, as desired.

Because POBSYS can retrieve data from a number of source files, it has the tools needed for real-time quality assurance checks. The software is able to compare current data against expected values or previous measurements and, thus, to reject erroneous or invalid data before it is permanently recorded. Such checks can be used to detect operations errors (e.g., by comparing a measured property of a component, such as its weight, against a value in a table listing component properties and identification) or to have the software restrict its own range of allowable actions by verifying the state of sensors and control points before initiating automated actions. This capability allows POBSYS to operate within a three-level safety system for fail-safe operation: software warning, software shutdown, and hardwired shutdown.

The computerized operating procedures developed for use with the FSS (OPRFSS) and with the DDS (OPRDS1 and OPRDS2 for separate and independent operations of the twin dissolver systems) are described in Sections IV.D and IV.E.

c. Stand-Alone Programs

Stand-alone programs in the project computer system served a variety of functions and were dispersed among all the DEC computers. The major programs in this category may be grouped into four broadly defined classes: (1) balance performance programs, (2) weighing routines, (3) data-entry programs, and (4) report generators. In general, each stand-alone program acquired data from a device (e.g., terminal or balance) interfaced to the computer or from data files previously generated by other programs, processed the data in a predetermined manner, and recorded new data in specific files for subsequent access. In many cases, the programs contained sequenced instructions for an operator to follow to ensure that the data required by the program would be entered in an appropriate order and would be complete. In such cases, the instructions were programmed in a "prompt-and-go" format, i.e., only one step was provided the operator at a time, and the next step would not appear on the terminal until the operator acknowledged that the current step was completed. Other programs, particularly the report generators, retrieved their own source data from files specified within the program and, once initiated, ran to completion without operator intervention, writing the output data into other specified files.

A few of the stand-alone programs are described below; these are described here because either they were used in many different areas of project operations (this is true of the balance programs, especially) or they are a representative example of their class. Many of the other programs are discussed in subsequent report sections that deal with the specific area of application of each program, e.g., operations or data management.

(1) Balance Performance Programs

Two of the most widely used stand-alone programs in the project computer system provided an objective assessment of balance performance prior to an operator using the balance for obtaining weight data.

The first of these programs, named BALQA (Balance Quality Assurance), provided accurate calibration, testing of linearity, and an estimation of the standard deviation of an individual weight measurement (object reading minus average balance tare). The operator is prompted to perform a specific series of weighings of the standard and test weights designated for use with the balance being tested. The program then compares calibration, linearity, and statistical parameters obtained from the weighings performed in the test with historical data and performance criteria (error limits) contained in a data file maintained in the computer for each of the 13 project balances. If the current test data are found to be inconsistent with the history of performance or limits to allowable error for the specified balance, the operator is instructed, by a terminal message, to carry out an appropriate action. Depending on the significance level of the inconsistency (based on the historical standard deviation of weighings with the balance), the operator may be instructed to repeat the test or to notify his supervisor that the balance is not functioning satisfactorily and is in need of adjustment or repair. When the test data meet the performance criteria specified in the program, the data file for the tested balance is automatically modified to update the historical statistics, to record current calibration information for the balance, and to record the date and time at which the testing was performed. The mere fact that the balance passed the tests specified in BALQA provides

considerable confidence that the balance performed at the same level in the current test as it did in previous tests, and that this level of performance is sufficient to meet project requirements on weighings made with the balance. By accessing information in the balance table (i.e., the computer data file for the balance in question), other programs in the project system could, in addition, verify that the balance was acceptable for use and retrieve calibration data needed to compute the most-accurate weights corresponding to acquired balance readings.

The other balance program used in the project was a truncated version of BALQA, known as BALCC or "Balance Calibration Check." It prompts the operator to carry out a single weighing of a specified standard weight and checks the result against an acceptance interval determined from the known weight of the standard and the statistics in the balance table. If the result falls outside the acceptance interval, the operator is instructed either to repeat the test or to run the BALQA program, depending again on the significance level of the discrepancy. The BALCC program provided a convenient check that the balance calibration had not drifted or balance performance deteriorated since the last BALQA test, without the operator performing the time-consuming multiple weighings required by the comprehensive tests of BALQA.

(2) Weighing Routines

Weighing routines were written to acquire, manipulate, compile, and store weighing data from specific project operations and were generally of the prompt-and-go type. They ranged in complexity from relatively simple programs for the out-of-cell weighing of sample bottles, sample cans, and transfer standards, to lengthy control programs that instructed the operator to perform specific sample manipulations, assigned sample numbers to objects to be weighed, and involved numerous loops and branches that executed on the basis of weighing results or operator options. These programs could be used independently of the POBSYS or could be called up by the POBSYS programs in executing specific steps in the DDS or FSS operating procedure. The most complicated of the weighing routines were those written to aid project analysts in preparing spikes needed for the mass spectrometric isotope dilution (MSID) assays of uranium in the dissolver solutions (program ALQ950 for making NBS 950a spikes and ALQ993 for making NBS 993 spikes), and the program, K3ALQ, that aided the aliquoting of dissolver solution samples in the analytical hot cell. These programs are described in detail in Documents C-0030-0288, -0289, and -0290 of Attachment 1.

(3) Data-Entry Programs

Not all of the project equipment was interfaced to the project computer system, and a means had to be provided for entering data from measurements using instruments that could not be interfaced into computer-file records. In addition, process planning information, such as shearing instructions or equipment-use designations, had to be translated from human-readable formats to computer files for use by the operations programs. The data-entry programs served these needs. These programs typically requested the operator to make specific keyboard entries of required information, checked the entries for consistency with expected format or expected range of value, and then wrote the entered data into an appropriate file. Programs used in entering data that had been manually obtained included the program FISGAS (used for entering results

of measurements with the gas mass spectrometers), RODGAS (used to enter pressure and temperature data obtained during puncture of the fuel rod plenum), and LOWMS (used for entering uranium and thorium isotope ratios obtained with the NBS TMS instrument). Programs for entering process planning information included P3 (the "Process Planning Program"), which created a Fuel Rod Information File (FRIF) that contained all of the information required by the computer system for the processing of an entire fuel rod, and HISLOG, a program used to maintain a set of files in which the history of each reusable piece of equipment in the DDS cell was recorded. Of course, the HISLOG files could also be automatically updated, when appropriate, by action of the operating system programs. The HISLOG files could be readily interrogated by the operator without interfering with process operations.

(4) Report Generators

Report-generating programs within the project system were created to perform the specific data processing steps that were needed to translate data collected in the operations into results formatted for convenient human perusal. In some cases, the report programs retrieved data from files generated in several different operations areas, combined the data, performed calculations, and wrote the calculated results into a report file that could subsequently be printed. In other cases, the report program merely reformatted information contained in one file and wrote it to another so that the information was more readable or easier to interpret during, for example, management review of a completed operation. Reports provided to BAPL on the destructive assay of each LWBR fuel rod were composed, in large part, of the output files from several of the report generators. These outputs included graphical as well as tabular formats for data presentation.

IV. OPERATIONS

Operations involved in the processing of each LWBR fuel rod during the EOL campaign were managed within a rigid framework, based in large part on the tools provided by the project computer system. The two major categories of operations management are process management and data management. Process management involves the planning and execution of process steps needed to physically accomplish the goals of the operations. It includes such considerations as specification of equipment for use in each operation, and steps required to ensure that specified operations are done in proper sequence. Data management involves the handling of information, both information coming to an operator regarding the work at hand and information acquired by the operator using process instrumentation.

Both the process-management and data-management requirements within the project operations were established prior to the onset of the EOL campaign through the use of detailed procedures, primarily computerized but sometimes only in written form. These procedures specified each step to be carried out, the sequence of steps in a particular operation, and the information needed for or expected from execution of a step. In establishing the computerized procedures, we made every effort to take advantage of the data retrieval, automated data acquisition, and decision-making capabilities of the computer system to aid the operator in carrying out each specified step. To satisfy specific formatting requirements of the computer and to simplify communication between operations areas, we established a standardized system of nomenclature for sample designation and computer file names on a project-wide basis.

In this report section, we describe the project operations, hoping to convey to the reader an understanding of the general management systems, an awareness of which operations were performed and how, and a basis for interpreting results presented in Section V. To achieve these goals, we shall not provide listings of individual step-by-step procedures (although these are available), but rather will focus on describing important process concepts and parameters; formulas used in computing key process results will be presented. Detailed listings of the shearing and dissolution procedures are available in Ref. 9.

In the discussion below, the order of presentation reflects, after some general considerations, the general sequence of operations in processing a fuel rod from initial receipt and unloading through reporting. Waste disposal is dealt with in Section III.A.6.

A. Sample Identification Format and Nomenclature

A comprehensive identification system for use in labeling samples that were generated during project operations was incorporated into all operating procedures and computer files. In this system, samples were identified by a string of characters that uniquely described each sample. Alternating fields of letters and numbers were used because this sequence is less prone to errors from transposition or misreading. The system is illustrated in the following example, which identifies a sample aliquot taken from the spiked BT for uranium analysis by 950a MSID:

<u>A</u>	-	<u>01</u>	-	<u>SB</u>	-	<u>1</u>	-	<u>U8</u>	-	<u>1</u>
Fuel Rod		Segment		Source Code		Sample No.		Analysis Code		Aliquot No.

The fuel-rod letter designation in the ANL system replaced BAPL's seven-digit serial number to simplify our internal sample identification procedure and provide easier labeling. Each rod was assigned a letter at the time of its receipt. When results for a rod were reported to BAPL, both the ANL letter designation and the BAPL serial number were specified.

Each segment was identified by a two-digit number following the rod letter designation; the numbers were assigned in the order in which the segments were excised from the rod, which was sheared into contiguous segments. For all rods, the gas plenum section was sheared and collected first; the plenum section was assigned the designation "-00-." The numbering continued consecutively, starting from "-01," for the immediately following fuel-bearing segments and continuing through to the last segment, a solid Zircaloy end cap or "stub" segment. The aluminum cans for collection of the sheared segments were labeled, using a permanent-ink marker, according to the identification numbers of the segments that they held, e.g., cans having segments from rod A were labelled A-00, A-01, etc.

When each segment was subsequently dissolved, three types of solution samples were removed from the dissolver system for analysis, and several other samples were generated, including BT decontamination samples and gas samples. Each of these solutions was assigned a specific source code indicative of where the sample came from or what it consisted of. The sample number following the source code specified where in the sampling sequence each particular sample from the indicated source was taken; in the example above, the sample was the first of several (usually four) removed from the spiked BT, the source code designation being SB.

If a given sample was divided into aliquots for several different or replicate analyses, information was added to the sample number to identify each aliquot taken. This was done by affixing a two-character analysis code that indicated the type of analysis for which the given aliquot was intended and a one-digit aliquot number that identified the individual replicate aliquot. The source and analysis codes used with various EOL samples are presented in Table 4.

The computerized operating procedures written for EOL operations were programmed to store data in association with rod, segment, and sample numbers consistent with this format. Thus, once familiar with the identification system, any operator or manager could relate the data to the appropriate sample at a glance. Furthermore, upon looking at the label on a given sample container, the operator could quickly discern where it came from, what had been done to it, and for what analysis it was intended.

B. Process Planning: The Fuel Rod Information File (FRIF)

The first step in processing each fuel rod at ANL was to provide information to the computer programs that control and carry out specific operations

Table 4. Source and Analysis Codes for EOL Samples

Sample Description	
Source Code	
DS	Secondary Dissolution Sample
UB	Unspiked Blend Tank Sample
SB	Spiked Blend Tank Sample
BT	Blend Tank Reflux Rinse
GS	Gas Sample
HU	Zircaloy Hulls
FR	Dissolver Filter
SS	Discretionary or Special Samples
 Analysis Code	
U5	^{235}U Spiked Uranium
U8	^{238}U Spiked Uranium
UU	Unspiked Uranium
CS	Fission Product ^{137}Cs and ^{144}Ce
ZR	Fission Product ^{95}Zr and ^{144}Ce
TH	^{230}Th Spiked Thorium

in the processing sequence. The information needed by the system included the rod identification number, details of the processing plan provided by BAPL for the rod in question, identification of equipment designated by project management for use in rod processing, and specification of rod-dependent process variables such as dissolution volumes for each segment sheared from the rod. This information was entered into the computer by means of the data-entry program, P3 (Process Planning Program), which creates a file, designated FRIFr (Fuel Rod Information File). (The lower-case "r" in the file name refers to the rod letter, e.g., A, B, C, etc. A file of the same format and name, but with the appropriate rod designation, was created for each rod to be processed.)

Running of the P3 program to create the FRIF was restricted to designated project personnel (Task Leaders). Some of the information in the FRIF was copied into the P3 program (by manual entry) from the shearing diagram provided by BAPL; an example of a shear plan is shown in Fig. 8. Other information was assembled from a filed data base by the program itself, using the rod serial number and tables of information that relate the serial number to the type of rod, its weight, linear density of fuel and cladding, etc. Still other information was specified by keyboard entry of equipment designations or values for variables. All entries were prompted by the program and were checked by the program for appropriate format and range of value before being accepted. An example of a completed FRIF is shown in Fig. 9, where FRIFB is shown.

SHEAR DIAGRAM

POWER FLATTENING BLANKET ROD TYPE 23
BOTTOM MOUNTED HIGH ZONE 70 INCH BINARY STACK

RELEASED FOR
SHEARING

DIAGRAM 2300711

CR
9/7/84

Fig. 8. Example of a Shear Plan Provided by BAPL

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Fuel rod #2606481 ANL ID "B"

Summary of general and FSS data:

Power Flattening Blanket Rod
 Nominal Diameter is 0.527 in.
 Top-mounted rod
 84.00-in. HZ Binary Stack Length
 Nominal Rod Weight is 3560. g

Seg no.	Length	Far bound	Near bound	Clad Wt (g)	Oxide Wt (g)	Total Wt (g)	Est. U content	Can size	Brush cup	M-3 bal
00	107.199	107.199	107.199	459.356	2890.245	3349.601	55.875			
01	10.993	107.199	96.206	47.820	300.878	348.698	0.125	6	BC-01	3
02	18.089	96.206	78.117	78.687	495.096	573.783	11.500	6	BC-02	3
03	17.500	78.117	60.617	76.125	478.975	555.100	11.500	6	BC-03	3
04	17.500	60.617	43.117	76.125	478.975	555.100	11.500	6	BC-04	3
05	17.500	43.117	25.617	76.125	478.975	555.100	11.500	6	BC-05	3
06	14.561	25.617	11.056	63.340	398.535	461.875	9.500	6	BC-06	3
07	9.456	11.056	1.600	41.134	258.811	299.944	0.250	6	BC-07	3
08	1.600	1.600	0.000	*****	*****	*****	*****	6	BC-08	3
91	*****	0.000	*****	*****	*****	*****	*****	6	BC-09	3
92	*****	*****	*****	*****	*****	*****	*****	6	BC-10	3
T1	107.199			459.356	2890.245	3349.601	55.875			

Shear blade to be used for shearing the rod: Blade 2 side B

Shear housing to be used for this rod: SH 0.527

Shear housing insert and orientation: 0.527 HI SIDE A

Cell M-3 transfer standards:

5000g transfer standard = TS5000X003
 2000g transfer standard = TS2000X003
 300g transfer standard = TS300X003

*** NOTE -- Starred or blank entries have not yet been entered for this rod

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Fuel rod #2606481 ANL ID "B"

Summary of general and DDS data:

Power Flattening Blanket Rod
 Nominal Diameter is 0.527 in.
 Top-mounted rod
 84.00-in. HZ Binary Stack Length
 Nominal Rod Weight is 3560. g

Seg no.	Diag no.	M-1 bal	Blend tank	950A spike	Filter hang 1	Filter hang 2	Gas cyl 1	Gas cyl 2	Thorex volume	Waste dest.
00	DS1	7	TK-08		FR-08	FR-08	SC-01	SC-01	1.0	WT-2
01	DS1	4	TK-01	50A-121	FR-01	FR-01	SC-01	SC-11	3.0	WT-2
02	DS2	4	TK-02	50A-303	FR-02	FR-02	SC-02	SC-12	3.0	WT-2
03	DS2	4	TK-03	50A-304	FR-03	FR-03	SC-03	SC-13	3.0	WT-2
04	DS2	4	TK-04	50A-305	FR-04	FR-04	SC-04	SC-14	3.0	WT-1
05	DS2	4	TK-05	50A-306	FR-05	FR-05	SC-05	SC-15	3.0	WT-2
06	DS1	4	TK-06	50A-302	FR-06	FR-06	SC-06	SC-16	3.0	WT-2
07	DS1	4	TK-07	50A-122	FR-07	FR-07	SC-07	SC-17	3.0	WT-2
08	DS*	*	TK-08		FR-08	FR-08	SC-08	SC-18	***	
91	DS1	*	TK-09		FR-09	FR-09	SC-09	SC-19	***	WT-2
92	DS2	*	TK-08		FR-10	FR-10	SC-10	SC-20	***	WT-2

Dissolution order for dissolver DS1:

B00 B01 B07 B05 B91

Dissolution order for dissolver DS2:

B02 B03 B04 B05 B92

Cell M-1 transfer standards:

5000g transfer standard = TS5000X004
 2000g transfer standard = TS2000X004
 300g transfer standard = TS300X004

*** NOTE -- Starred or blank entries have not yet been entered for this rod

This file was last edited on 21-AUG-84 at 13:25:24
 F.J.MARTINO, CHANGED B-04 WT DESTINATION.

Fig. 9. Example of a Completed Fuel Rod Information File (FRIFB)

Because of the importance of the FRIF in setting parameters for operations on each rod, special precautions were taken to ensure its integrity. One of these precautions was the restriction of access to the P3 program mentioned above. The program could be run only by personnel whose password matched a list of privileged passwords. As further precautions, the version of the FRIF that was accessed by the computer during operations was stored in a binary format that could not be readily edited except through the P3 program, and the program generated a log of any editing that was done after the original FRIF was created (see example in Fig. 9). The formatted version of the FRIF shown in the example was created from the unformatted (binary) original by a separate program written for this purpose, which could be run by any project member. In this way, all project personnel were given access to the information in the FRIF, but only authorized personnel could change the information.

Because much of the input to the P3 program required manual data entry by an individual, a formal procedure for review of the FRIF content was established whereby two task leaders other than the one who entered the data were required to verify the correctness of the information and to give a signature record of their review. A signed copy of each FRIF was kept on file in the project manager's office.

C. Receipt and Unloading of Fuel Rods

Single- and multiple-rod shipments were transported to ANL in a 16-ton heavily shielded WAPD-40 shipping cask. Each rod within the cask was packaged in an individual secondary container specific to the rod type; most shipments also contained empty secondary containers as dunnage in the shipping cask.

Unloading of each shipment was carried out at the loading dock of the Shielded Cell Facility according to a written procedure, which was designed to ensure personnel safety during operations with the highly radioactive rods, as well as to preserve the integrity of the rods being unloaded. No significant problems of any kind were encountered during any of the cask unloadings. A list of the shipments received at ANL during the EOL campaign and the rods contained in each shipment is shown in Table 5.

Each rod received at ANL remained inside its secondary container until being transferred into the FSSF at the time processing of the rod was started. In a few cases, a rod was moved directly into the FSSF as part of the cask-unloading operations. Most often, however, the rod was placed in the storage cell (see Section III.A.1) and was transferred to the FSSF at its turn in the processing sequence. In keeping with BAPL's wishes, we processed the rods one at a time, with no rod entering the FSSF until all segments from the previous rod had been transferred to the DDS facility. In addition, no segments were transferred to the DDS until all segments from the previous rod were fully processed and the samples transferred to the ACL.

D. Operations in the Full-Scale Shear Facility (FSSF)

Operations performed in the FSSF for each LWBR fuel rod included the following: (1) visually inspecting the rod and photographing it as deemed appropriate; (2) removing the rod's mounting tang; (3) weighing the whole rod

Table 5. Fuel Rod Shipments Received at ANL during the EOL Campaign

Shipment	Date Received	Number and Types of Rods (ANL Letter Designation)
1	August 13, 1984	Three PFB Rods (B, C, D)
2	September 10, 1984	Three PFB Rods (E, F, G)
3	November 5, 1984	One Reflector Rod (H)
4	December 17, 1984	Three SB Rods (I, J, K)
5	January 28, 1985	One SB Rod (L)
6	March 25, 1985	Five Seed Rods (M, N, O, P, Q)
7	September 23, 1985	One Reflector Rod (R)

(both before and after plenum puncturing); (4) puncturing the plenum end of the rod, to determine the quantity of gas contained in its void volume and provide gas samples for analysis; (5) measuring the rod length and surface temperature; (6) shearing the rod into contiguous segments; (7) weighing the collected segments; and (8) weighing the brushes used for intersegment cleaning of the shear blade. Each of these operations was performed in accord with the computerized operating procedure OPRFSS. Through use of the POBSYS software system, OPRFSS called up terminal screen displays that instructed the shear operators to perform specific manipulations of equipment or materials, acquired data from instruments interfaced to the computer system, and executed semi- or fully-automated portions of the operations. Use of OPRFSS gave full replication of the entire rod shearing process, which comprised some several hundred steps.

The computer system also continuously monitored specified sensors in the FSSF and alerted the operator to any conditions outside specifications by means of an alarm message on the terminal accompanied by an audible alarm. Intermediate calculations involved in the data-taking process were carried out automatically by overlay (slave) routines called for by OPRFSS, and results were presented on the terminal screen for operator acceptance. Checks on data quality and acceptability were also built into the computer programs and made automatically by the computer as data were taken. For example, balance tare readings would not be accepted by the OPRFSS if they fell outside a small interval around zero, and weights of empty sample cans were not accepted if they fell outside a specified range of weights deemed optimum for the subsequent dissolution operation. When such rejections of data by the computer occurred, a displayed message to the operator relayed the necessary recovery action to be taken, e.g., adjust the balance tare, select another can. Progression from one step in the operating sequence to the next was initiated by the computer system only after the operator indicated, by means of a sign-off entry of his or her initials, that the current step was satisfactorily complete.

Records of the FSSF operations for each fuel rod were generated in several different formats, which provided redundant records of critical information and made particular data items available in a convenient format for management review or for computer access during subsequent operations that required retrieval

of the stored data. A detailed record of the operations performed under the control of OPRFSS was maintained in the computerized process log file. For each step in the OPRFSS procedure, entries were automatically made in this log, which included (1) the step number, (2) the time and date of execution, (3) the identity of the operator who signed off the step, (4) data acquired during step execution, (5) alarm messages active during the step, and (6) comments entered by the operator regarding the operation. To provide a hardcopy backup of the process log information, each entry made to the process log computer file was simultaneously printed at the FSSF work station printer.

The volume of information contained in the process log file did not lend itself to efficient computerized searches to retrieve data needed for later operations or reports. To simplify such searches, selected data items were simultaneously stored in a summary data file, which contained the analytical information obtained during the shearing operations, and in specialized data files that contained data acquired during specific operations or data that related to a specific report to be generated after the rod was processed. In addition, a manual logbook was maintained at the FSSF work station, wherein the operators could note observations made during the day-to-day performance of their duties in the work area. Such notes included, for example, observations made during scheduled or unscheduled maintenance of the equipment or notes regarding the appearance of the fuel rods. Each operator was also encouraged to record any suggestions that might improve the clarity of instructions presented in the OPRFSS terminal displays, the execution of manual procedure steps, or any other facet of the FSSF operations, so that appropriate management consideration of the suggestions could be made.

In the following subsections, major operations within the OPRFSS procedure are described individually, more or less in the order in which they were performed for each rod. Key features of the operations are listed in most cases, but the descriptions are not intended to be exhaustive. Any reader interested in the step-by-step execution of the FSS operations should refer directly to the OPRFSS procedure documentation.⁹ Equipment in the FSSF was described in Section III.A.3.

1. Preliminary Operations

Before the fuel rod was moved into the FSSF hot cell, a number of operations were carried out to check performance of the equipment and to provide materials needed for shearing a given rod; these operations are discussed in Sections a through d, below.

a. Tooling Assembly and Feed System Calibration

The first operations performed in preparing to shear each fuel rod were the assembly and installation on the FSS of the appropriate, clean shear tooling for the rod to be processed (cleaning of the tooling is discussed in Section IV.D.11) and the installation of an appropriate tip on the sensing shaft of the rod-end locator (REL). The tooling was selected, per guidance from the FRIF, from among the dies stored in the cell and was assembled with a new O-ring in the rod-entry-port sealing mechanism prior to being installed on the shear base. The REL tip conformed to the bottom-end configuration of the rod. A flat tip was used for top-mounted rods, and a specially machined tip (to accommodate the mounting-tang protrusion), corresponding to the rod size, was used for bottom-mounted rods.

After these installations, an operations check of the shear hydraulics and tooling was undertaken. When satisfactory mechanical operation of the shear was verified, the feed/measurement system was calibrated as described in Section III.A.3.d.(4). The precision gauge rod used for the calibration procedure was also used at this time to establish values for two variables that enter into subsequent calculations. These variables are (1) the carriage position when the back end of the gauge rod is at the REL reference position and the lead end is in contact with the forward stop, and (2) the distance between the shear plane and the forward stop, which corresponds to the distance that the fuel rod must be retracted to begin cutting operations.

Operationally, these tasks were accomplished as follows. First, the gauge rod was placed on the shear bed with one end in contact with the lowered shear blade; at this time, the rod was held in place with three hold-down positioners. Then, the computer calculated the true gauge rod length (i.e., corrected for thermal expansion) and displayed the resulting value for the operators. The rod temperature was established as the average of readings from RTD sensors that were mounted on selected hold-down positioners such that the sensors contacted the rod when the positioners were lowered. The carriage (see Fig. 2) was then manually indexed (following computer instructions) to the back end of the gauge rod, until the temperature-corrected readout for the absolute shaft transducer (AST) corresponded to the temperature-corrected gauge-rod length (to the nearest 0.0005 in.), and the REL readout indicated that it was near its midrange of travel. The REL digital readout was then manually set, if necessary, to a value of 0 ± 0.005 in. This setting provided nearly full REL-range sensing of rod-end displacements in either direction (the REL readout displays the sign as well as the magnitude of displacement, up to ± 0.050 in., from its zero position). Once established, the REL set point was not changed for the remainder of operations with the particular installation of tooling.

To verify proper operation of the automated FSS control system and to provide a representative average REL reading for use in computing the REL reference value (see below), we retracted the carriage 1.000 in. from the rod end and returned it to its original position, in automated mode, three separate times. For each trial, the AST and REL readings were automatically recorded. To be acceptable to the computer system (and the operator), all three AST values had to be identical, and the three REL readings had to fall within 0.0001 in. of one another. These tight tolerances were met consistently and are indicative of the precision with which carriage positions on the feed system could be established and measured. The average of the three REL readings and the temperature-corrected AST value were stored by the computer for use in determining the distance between the shear plane and the forward stop, as described immediately below. The computer also calculated the REL reference value by determining the value which, when taken as the reference point, would correspond to an REL displacement equal to the difference between the temperature-corrected AST value and the true gauge-rod length. This REL reference value was also stored by the computer for use in subsequent operations.

For the next step, the shear blade was raised and the gauge rod was pushed forward through the exit bore on the far side of the shear tooling, such that the rod was in contact with the forward stop. The carriage was then indexed to the back end of the gauge rod, such that the REL read its reference value to ± 0.0005 in., and a set of three AST and REL readings was obtained in

automated mode. The temperature-corrected AST average (AST_{FS}) and the average REL reading (REL_{FS}) were stored in the computer for later use in the rod length calculations (see Section IV.D.5). These values were also used, together with the AST and REL values measured with the gauge rod in contact with the shear blade (AST_{SP} and REL_{SP} , respectively) to calculate the distance between the shear plane and the forward stop (D_{SP-FS}). This distance is equal to the difference between the carriage positions for the two gauge-rod locations, i.e.,

$$D_{SP-FS} = (AST_{SP} + REL_{SP}) - (AST_{FS} + REL_{FS}) \quad (IV-1)$$

where SP indicates shear plane, and FS indicates forward stop.

Acceptance of the D_{SP-FS} value by the shear operator completed the shear calibration. The gauge rod was then returned to its in-cell storage rack, and the pre-processing work continued.

b. Balance Tests

Upon completion of the shear checkout and calibration, quality assurance checks of the three in-cell balances were performed. Because of the importance of these balances in meeting requirements regarding fuel-rod and segment weights, criteria for acceptable performance of the FSSF balances were more demanding than for any other balances used in the project.

Three types of standard weights were used for calibrating and verifying the calibration of each balance: (1) the in-cell standard weights associated with each balance (see Section III.A.2.d); (2) the project reference standards, which were individually calibrated by NBS and kept outside the cell; and (3) transfer standards. The transfer standards consisted of lead shot in a polyethylene bottle and were weighed outside the cell on a balance calibrated against the project reference standards. They were then weighed inside the cell on a corresponding balance calibrated with the in-cell standard weights. Agreement between the out-of-cell and in-cell weights of the transfer standard indicated consistency between the in-cell and project reference standards, and was required for acceptance of the in-cell balance calibration. Discrepancies between the in-cell and out-of-cell transfer-standard weights were to be interpreted as an indication that the integrity of the in-cell standard weights had been compromised, e.g., the weights were dirty or damaged. However, no differences of consequence were noted during the campaign.

During the balance performance tests preparatory to processing each fuel rod, each balance was first calibrated with the in-cell standard weights, and then was tested for linearity, precision, and stability by means of the BALQA program [see Section III.C.2.c.(1)].

The out-of-cell weighings of the individual transfer standards were done under the control of a stand-alone computer program (TSOWR, "Transfer Standard Original Weighing Routine"). This program prompted the operator through the weighing steps, read the balance, and stored the weighing data in a file, TSxxx.DAT, created specially for each transfer standard. (The transfer

standard to be used for each balance test was specified in the FRIF.) When the transfer standards were weighed inside the cell, another program, a slave to OPRFSS and designated TSFWR ("Transfer Standard Final Weighing Routine"), was used. Besides carrying out the same functions as TSOWR, TSFWR compared the in-cell and out-of-cell weights and advised the operator concerning the significance of any differences observed. For the in-cell weights to be judged acceptable, the average of three in-cell and three out-of-cell weights had to agree within the readability of the balance used for the weighing. (The in-cell and out-of-cell balances were identical models, except that the in-cell units had remote electronics, as described in Section III.A.3.a.) The results of the weighings for each transfer standard (each was used only once) were recorded in the TSxxx.DAT file, along with the judgment made by the computer regarding acceptability of the results, and were kept as part of the project's permanent computer records.

Each program that used the FSSF balances to acquire weighing data, in subsequent operations, checked the appropriate balance records and would not run unless the prescribed balance had passed BALQA within the previous 24 h and BALCC in the previous four hours. If too long a time had elapsed since the previous balance test, OPRFSS automatically called up the appropriate test program to run before operations were continued.

c. Weighing of Aluminum Sample Cans

The next operation performed was preparation and weighing of the aluminum cans used to collect sheared segments; all weighing data were again automatically taken and filed by the computer. In this operation, the proper number of cans with caps was first selected from a batch that was previously degreased using Freon solvent and inspected for cleanliness. Next, a permanent-ink marker was used to separately label each cap and can with the appropriate rod letter and segment number. Each can plus cap was then weighed to 0.001 g on the appropriate out-of-cell balance under control of the program SCOWR ("Sample Can Original Weighing Routine"); this program computes the empty can weight as the average of three separate weighings. Each can and cap were weighed together as a set to provide the most precise total weight of each empty can. They were weighed again, separately, to allow replacement of one part or the other in the event one item was damaged during later operations. The program SCOWR contained provisions for weighing the replacement part on an as-needed basis. The results of the out-of-cell can weighings were stored in a computer file by this program for later comparison with in-cell weighing data.

After the sample cans were transferred into the cell, each was again twice weighed in triplicate, both can and cap together and can and cap separately. An overlay routine in OPRFSS was used for this step. The individual can and cap weights were automatically recorded in the computerized OPRFSS process log and in the shearing data summary file, along with the difference between the average in-cell and out-of-cell weights.

d. Weighing of Brush Assemblies

Each fresh brush assembly (consisting of a metal brushcup and nylon-disk insert) that was designated in the FRIF for intersegment cleaning of the shear blade was weighed inside the cell to 0.001 g. Only one weighing of each brush assembly was made, since these data were used only for purposes of

process control and evaluation and did not enter into calculations that affected key results of the destructive assay. The brush-assembly weights were taken under computer control and were stored in a file by the computer for use in subsequent material balance calculations.

2. Visual Examination and Identification of the Fuel Rod

Processing of the fuel rod began with its unloading from the secondary container and its insertion into the FSSF alpha enclosure and onto the shear bed. The rod was then rolled from the bed onto an aluminum support trough (cradle), which was normally stored in a recess in the shear bed. The cradle provided support for the rod and prevented its bending during operations that required manual manipulations of the rod or moving of the rod from place to place in the cell, e.g., during weighing operations. After the rod was in place on the cradle, the assembly was moved with manipulators to a position in front of the cell viewing windows. In accord with project requirements, care was taken to keep the plenum end of the rod slightly elevated during all such moves. This precaution was intended to prevent fuel fragments from shifting into the plenum region, since such a shift could confound comparison of the destructive analysis data with the nondestructive assay by BAPL.

At the viewing windows, the rod was visually inspected for any unusual blemishes on the cladding, and the rod serial number was verified. If any notable features were observed on the rod, these were noted in the FSSF logbook and photographed at 2X magnification. Following this inspection, the cradle containing the rod was moved to the enclosure floor in preparation for removal of the rod-mounting tang with the pneumatic bolt cutter. The tang was removed to facilitate measurement of the rod length. After the tang was cut off (and discarded to waste), the rod was returned to the shear bed in preparation for weighing.

3. Weighing the Fuel Rod

Each fuel rod was weighed twice, once before and once after the rod was punctured for release of the plenum gases. Both weighings were carried out in the same manner with the weighing system described in Section III.A.3.a.

Weighing the fuel rod first required determining the tare for the aluminum cradle. The empty aluminum cradle was inspected for cleanliness and placed on the pneumatic lifting yokes of the fuel-rod weighing system. The empty cradle was then weighed three separate times to 0.1 g.

After the empty cradle was weighed, it was moved from the weighing system to the shear bed, and the fuel rod was rolled onto it. Taking care to not allow the plenum end to descend below horizontal, we then moved the assembly to the weighing system and weighed it three times. Finally, the rod was returned to the shear bed, and the empty cradle was weighed three more times. The fuel rod weight was computed as the difference between the average of the three rod-plus-cradle assembly weights and the average of the six empty cradle weights. The computer program that controlled these weighings recorded each individual cradle or assembly weight, as well as the calculated rod weight.

4. Puncturing of the Rod Plenum

Because the pressure and temperature transducers in the plenum-gas collection system were not interfaced to the project computer, the plenum puncture operations were performed according to a separate written procedure, and data were recorded manually by the operators on a standardized data record form. The operators were, however, instructed by the OPRFSS program to refer to this written procedure at the appropriate point in the process sequence. After the completion of FSSF operations for each rod, the puncture data were manually entered into the computer for calculation of results by means of a data-entry program, RODGAS. This program created, for each fuel rod, a file that could be accessed for further manipulation of the data by other programs, especially those programs that generated reports on the plenum gas measurements. An example of a file produced by RODGAS is given in Fig. 10.

Before the plenum was punctured, the support cradle was used to move the rod to the puncture station, and the saddle-valve puncture die was clamped around the rod at an appropriate position determined from neutron radiographs of the rod plenum end, which BAPL provided for each rod. The puncture position was selected to avoid the ceramic fuel stack and to facilitate penetration of the piercing pin through the rod cladding. The gas collection system and the interconnecting tubing up to the puncture die's gasketed seal at the surface of the rod were evacuated. Next, the gas collection system was leak-tested by isolating it from the vacuum pump and monitoring the pressure transducers for a period of five minutes; if no significant pressure increase was observed during this period, the system was judged leak-free. The five-minute pressure readings on each transducer were recorded in the RODGAS file as a record of the test results. The temperature of the fuel rod was measured with a portable RTD sensor held against the rod surface.

Next, the two calibrated expansion volumes in the gas-collection system were closed off, and the cladding was punctured. The piercing pin was then retracted slightly to release the gases from the rod, and the released gas was expanded into the first calibrated volume (377.0 cm³). After the gas pressure in this volume had stabilized, the pressure and temperature of the gas were recorded, and the gas was expanded into the second calibrated volume (318.9 cm³). Again, the temperature and pressure of the gas were recorded. Finally, the gas was expanded into each of two gas-sampling cylinders to provide samples for the subsequent mass-spectrometric analysis (see Section IV.F.2), which was used to determine the gas composition and fission-gas isotopy. Pressures and temperatures were also recorded after each of these expansions.

The valves on the sampling cylinders were closed, and the cylinders were removed from the system and submitted for analysis. The two samples were designated r-00-GS-1 and r-00-GS-2, where the lower-case r stands for the rod letter identification.

The gas that remained in the collection system was later diluted with cell-ventilation air and slowly released. The quantity of ⁸⁵Kr in the released gas was measured by integrating the signal from the in-line stack monitor over the period when the release took place. This information was used in assessing the validity of the stack-monitor data obtained for gases released during the shearing of individual segments from the rod, as discussed in Section V.G.

PLENUM PUNCTURE GAS MEASUREMENT

FISSION GAS MEASUREMENT

ROD NO. 2606481
 ROD TYPE PFB
 ROD TEMP. 31.1

DATE 8-18-84
 TIME 0845

Step	VOLUME 1		VOLUME 2	
	ABSOLUTE PRESSURE mm Hg	TEMP. C	ABSOLUTE PRESSURE mm Hg	TEMP. C
4.1.3.3.3 Initial Pressure	-0.8	29.2	-1.4	29.1
4.1.3.3.4 Five min pressure	-0.6	29.1	-1.3	29.1
4.1.3.4.7 Release to Vol. 1	44.6	29.2	-0.7	29.1
4.1.3.4.8 Initial Vol. 2			-0.7	29.1
4.1.3.4.10 Release to Vol. 2	25.0	29.2	25.0	29.2
4.1.4.5.3 After Sample #1	21.0	29.2	0.0	0.0
4.1.4.5.15 After Sample #2	18.9	29.2	0.0	0.0

PERFORMED BY: J. HOH

Data Summary: FIRST EXPANSION VOLUME: 377.0 cc
 SECOND EXPANSION VOLUME: 318.9 cc
 PRESSURE AFTER RELEASE TO VOL.1 45.2 Torr
 PRESSURE AFTER RELEASE TO VOL.2 25.7 Torr
 AVERAGE SYSTEM TEMPERATURE : 29.2 C

Results:	Description	Value	Estimated Uncertainty (One-Sigma)
	PLENUM VOID VOLUME	41.4 cc	6.1 cc
	PLENUM GAS PRESSURE	456.8 Torr	61.1 Torr
	PLENUM GAS QUANTITY	0.001003 moles	0.000012 moles

Fig. 10. Example of RODGAS Data File (RODGASB.DAT)

When the pressure and temperature data recorded during the foregoing operations were entered into the RODGAS program, the program not only formatted and stored the information, but also performed calculations to (1) correct for the non-zero baseline pressure readings that are characteristic of the capacitive manometers used in the system, (2) to average appropriate redundant information (e.g., after expansion into the second calibrated volume, both pressure transducers are reading the same pressure and the two readings could be averaged), and (3) to derive a value for the total number of moles of gas collected from the rod

void volume. These last calculations were done with formulas that assumed ideal-gas behavior for the collected gas, as follows.

First, an approximate value for the rod void volume, V_x , was calculated from the known volumes of the expansion volumes, V_1 and V_2 , and the corrected values for the measured pressures on expansion, P_1 and P_2 . The use of two expansion volumes in the system made this calculation possible. The calculated rod void volume is only approximate because it arises as a small difference between large numbers and is subject to large uncertainties. From the ideal gas law, assuming constant-temperature expansion from the rod to the first and then to the second expansion volume, one has the relationship

$$P_2 (V_1 + V_x) = P_1 (V_1 + V_2) \quad (\text{IV-2})$$

Solving for V_x , one gets the formula

$$V_x = \frac{P_1 V_1 - P_2 (V_1 + V_2)}{(P_2 - P_1)} \quad (\text{IV-3})$$

Using the calculated value for V_x , one can determine the product $P_x V_x$ (P_x is the pressure of the gas in the rod void volume) from

$$P_x V_x = P_2 (V_x + V_1 + V_2) \quad (\text{IV-4})$$

which is analogous to Eq. IV-2. Because V_x is small relative to V_1 and V_2 , the large relative uncertainties in V_x contribute only small relative errors to the value for the $P_x V_x$ product. Finally, the total moles of gas present in the rod void volume, n , was calculated from

$$n = \frac{P_x V_x}{R T_x} \quad (\text{IV-5})$$

where R is the gas constant (0.0821 L-atm/mol-K), and T_x is the rod temperature in K.

The RODGAS program also (1) calculated a value for P_x by dividing the $P_x V_x$ product by V_x and (2) estimated uncertainties in all the calculated values by propagation of the measurement uncertainties associated with calibration of the expansion volumes, determination of the pressures, and determination of the temperatures that entered each calculation. Because the large uncertainties that accrued to the P_x and V_x results made them of only marginal interest, these values were not reported to BAPL; only the total moles of gas in the rod void volume was reported.

5. Measurement of Fuel Rod Length

When the post-puncture weighing of the fuel rod was completed, the rod was positioned on the shear bed for measurement of its length. With the shear blade retracted (and a safety block installed to prevent the blade from being lowered) and the shear-housing bore plug removed (see Fig. 3), the rod was indexed forward through the shear-housing bore until its lead (plenum) end was in contact with the face of the fixed forward stop.

The rod was held straight and flat during the length measurement by the pneumatically operated hold-down positioners on the shear feed system. The rod surface temperature at three locations on the rod was measured by means of RTD temperature sensors on three of the positioners; with the lead end of the rod in contact with the forward stop, these RTDs were located approximately 14.75 in., 26.25 in., and 88.25 in. from the lead end of the rod.

In determination of the rod length, the carriage on the shear feed system was indexed to the back end of the rod until the REL digital display read the reference REL value to ± 0.0005 in. (See Section IV.D.1.a. for determination of the REL reference value.) The computer then initiated a sequence of moves, whereby the carriage was retracted 1.000 in. and repositioned at the rod end three times, with the AST and REL values being read each time the carriage was repositioned at the rod end. It then displayed the three pairs of readings and an average AST and REL value for operator acceptance. If the operator accepted the readings and they met the test criteria of identical AST values and REL values within 0.0001 in. of one another, the computer read the RTD sensors in the shear machine base and on the hold-down positioners and then calculated, from these readings, an average machine-base temperature and average rod surface temperature.

Using the average machine-base temperature, the computer calculated a temperature-corrected AST value with the carriage at the rod end, AST_{RE} . The length of the fuel rod, L_{FR} , was then computed from the relationship

$$L_{FR} = (AST_{RE} + REL_{RE}) - (AST_{FS} + REL_{FS}) + L_{GRT} \quad (IV\ 3)$$

where REL_{RE} is the average REL reading with the carriage positioned at the rod end, L_{GRT} is the temperature-corrected length of the gauge rod used in determining AST_{FS} and REL_{FS} , and the other parameters are as defined for Eq. IV-1.

Each individual AST, REL, and temperature value, the calculated averages, and intermediate and final results were stored in the FSS computerized log, as well as in the shearing data summary file.

6. Shearing the Fuel Rod into Segments

The procedure used in shearing each fuel rod was developed to accomplish three primary objectives: to ensure (1) the precise location of segment boundaries, both in placing the boundaries at positions specified in BAPL's shear plan and in measuring the location of the boundary as actually achieved; (2) complete breakout of the ceramic fuel from the rod cladding and comminution of the fuel for efficient dissolution; and (3) maximal recovery of the sheared

materials with minimal carryover (cross-contamination) of material from one segment to the next. To meet the first objective, the rod was clamped in the carriage clamp jaws and the rod position was monitored by the computer using the sum of the temperature-corrected AST value and the REL displacement from its reference value as the control parameter. In effect, the location (relative to the rod reference end) of each shear cut made on the rod was monitored by measuring the length of rod that remained between the REL and the shear plane. To accomplish the second objective, each fuel-bearing segment was sheared from the rod as a series of short cuts, each approximately one-half the rod diameter in length. During characterization testing of the FSS, this cut length was found to yield relatively complete breakout of the fuel from the cladding and a favorable particle-size distribution in the sheared fuel. The third objective was met by employing slow-speed shearing (2.0 in./s movement of the shear blade) to minimize ejection of small fuel particles from the blade housing during each shear stroke and by cleaning the shear-blade bottom between segment cuts.

The entire shearing operation for each rod was carried out under full computer control. To begin the operations, the fuel rod (its lead end still in contact with the forward stop from the length measurement) was clamped in the carriage clamp jaws. The computer then retracted the rod a distance equal to the distance from the shear plane to the forward stop, D_{SP-FS} , plus one inch. This action positioned the lead end of the rod 1.000 in. from the shear plane. With the rod in this position, the operators installed the shear-housing bore plug and then checked the shear hydraulic system. The rod was positioned in this way so that the operators did not have to be concerned about damaging the rod end while carrying out these preliminary operations.

One of two procedures was used in clamping the rod for retraction, depending on the length of the fuel rod being processed. If the rod was classified as a short rod (<111 in. long), the carriage was indexed to the back end of the rod such that the REL read its reference value to ± 0.0005 in., and the rod was clamped in this position. For longer rods (all LWBR fuel rods except the reflector rods fell into this category), the REL was pivoted on the carriage such that it did not engage the rod end, and the carriage was indexed 13.000 in. forward from the REL reference position before the rod was clamped. This positioning of the carriage relative to the rod end was necessary because insufficient length was available on the feed bed for the back end of the carriage when the carriage was positioned at the rod end and the rod was retracted to its position on the feed side of the shear plane. The computer program that controlled the shearing operations selected the appropriate clamping procedure based on the measured length of the rod at hand and provided instructions for the operators to follow in clamping the rod. In the case of the long rods, the rod was reclamped at the rod end, with the REL engaging the rod, after the first (i.e., plenum) segment was sheared from the rod.

A repetitive cutting cycle was used to shear each segment from the rod in its turn. Operations for each segment consisted of the following basic steps, together with a few computer-initiated branching steps, as indicated.

- (1) The operators were instructed to select (per FRIF designation) and install the appropriate sample can and brush assembly in the sample collection station and to put the can in position under the

shear. The computer also verified that a can was in position by checking the "can-up" sensor on the collection station every time it prepared to lower the shear blade for a cut.

- (2) The computer calculated a plan for the segment to be sheared, based on the desired segment boundary location and the nominal cut length that were specified in the FRIF for this segment. The cut-plan calculation was based on the premise that the distance between the starting rod position for the segment and the desired boundary-cut position could be almost exactly expressed in terms of some specific number of cuts at each of two cut lengths that differed by only 0.001 in. The calculation, thus, computed the two cut lengths and the number of cuts at each length that would be required to reach the segment boundary. By establishing a boundary condition such that the total number of cuts at either cut length would equal the number of cuts required to span the segment length if the nominal cut length were used, the computer was able to constrain the magnitude of each of the two calculated cut lengths to a small interval (± 0.005 in.) about the nominal value. This relatively complicated approach to laying out the shearing of each segment had two important advantages. First, the computer could readily keep track of progress toward reaching the segment boundary by merely counting the number of cuts made at each calculated cut length as the segment was sheared; second, all cuts were close to the optimum length, and it was never necessary to make a very long or very short cut to compensate for the segment length not being an exact multiple of the nominal cut length. Details of the cut-plan calculations are given in Appendix A. The values of variables used in the calculations for each segment and the results of the calculation were automatically recorded in the FSSF computerized log and in the FSS summary data file. The results were also displayed on the work station terminal for operator acceptance.
- (3) Once the operator accepted the cut-plan parameters by a sign-off entry, the computer proceeded into a fully automated feed/cut cycle to shear the segment. In each repetition of the cycle, the following operations were performed: (1) the shear blade was raised, (2) the fuel-rod seal at the shear tooling entry port was opened, (3) the carriage was indexed forward by an amount equal to the appropriate length of cut and the fuel-rod seal was closed, (4) the surface temperature of the fuel rod at the location of the hold-down positioner nearest the shear plane and the cell ambient temperature were read, and (5) the shear blade was lowered, making the cut.

Data acquired during each operation were automatically recorded in the computerized process log and in several specialized computer files. One of these files contained a list of the rod positions, rod surface temperature readings, and cell ambient temperatures that were measured during operation (4) above and

was subsequently used to prepare a plot of rod surface temperature vs. position on the rod. This surface temperature profile was of interest to BAPL and was included in ANL's report for each rod processed (see Fig. 11 for a sample plot).

Fig. 11. Sample Plot of Rod Surface Temperature vs. Position on the Rod

Another set of files generated during the cycle contained blade position vs. time data acquired from the shear-blade position sensor (LVDT) for every cutting stroke. Through running a special, stand-alone program, we could use these files to graphically represent the shear blade movement during any individual shear cut. The files thus provided a useful tool for examining shearing characteristics for different rod types and different sections of a given rod. Examples of blade position vs. time plots are shown in Fig. 12.

The feed/cut cycle was repeated by the computer until the next-to-last cut for a given segment had been made. At this time, a

Fig. 12. Sample Plots of Blade Position vs. Time
(top: cut through fuel segment; bottom:
cut through solid Zircaloy)

branching step in the cycle was initiated so that the AST reading required to position the rod exactly for the desired boundary cut location could be calculated. The appropriate AST reading was computed as that value which, when corrected for the current average machine-base temperature and added to the REL displacement from the REL reference point, would give the segment boundary cut location called for in the shear plan. For the first segment boundary cut for long rods, where the REL was not in contact with the rod end, the calculations presumed that the REL displacement was negligible. The 10.000-in. advancement of the carriage from the rod end was also accounted for in this case.

The calculated AST reading for the boundary cut was displayed for the operator's acceptance, and then the rod was automatically indexed to the calculated position and the cut was made. Next, the computer read the REL, AST, and average machine-base temperature and calculated the actual, as-cut location of the segment boundary. For the first segment of a long rod, the operators were instructed to reposition the carriage at the rod end and to replace the REL after the actual boundary location was calculated. In general, any discrepancy that might have occurred between the desired and actual cut locations could be attributed to movement of the rod during the boundary cut (see Section V.B.1 for a discussion of rod movement during shearing of the LWBR fuel rods).

Values for all of the variables that entered into the boundary-cut-position calculations, both for the desired and actual locations, and the results of the calculations were automatically recorded in the computerized process log and in the FSS summary data file.

Two other computer-initiated branching steps were built into the segment shearing program. One of these was initiated when the carriage reached the position on the feed bed at 22 in. from the shear plane. It called for the operators to install an appropriate-diameter (corresponding to rod type), 22-in.-long push rod behind the fuel rod end and to clamp the push-rod end in the carriage clamp jaws. Use of a push rod in shearing the last section of the fuel rod was necessary because the carriage could not be indexed far enough forward to position the rod end at the shear plane because of the physical length of the carriage. In setting up the shearing procedure, we chose the 22-in. length of the push rod to avoid position measurements along the last 18 in. of the ball-screw lead end. This portion of the ball screw was found, during a laser-based calibration test of the FSS (see Attachment 3), to show a relatively large error for the length equivalent of the AST readout. The error was due to a dislocation in the lead of the ball-screw threads. The forward end of the push rod for all diameters had a slightly concave face to ensure colinear engagement with the fuel rod end. Using the change in AST and REL values that accompanied the push rod's installation, the computer automatically determined the apparent length of the push rod and compensated for the corresponding

carriage displacement in making all subsequent calculations of rod position and cut location.

The remaining branch in the segment shearing program was initiated when the last shear cut through the solid Zircaloy end cap (stub end) of the rod had been made, and only a short (usually 0.400-in.) section of the stub remained on the shear bed. At this point, the feed/cut program raised the shear blade, indexed the carriage forward 0.500 in. so that the push rod could push the stub end section into the sample collection can, and retracted the carriage 3.000 in. to bring the push-rod end away from the shear plane before lowering the blade.

- (4) When the shearing of each segment was completed and the results of the boundary location measurements for the segment were stored, the computer instructed the operators to cap the sample can containing the sheared segment, remove the can from the sample collection station, and then set the full can aside for later weighing.
- (5) The operators were next instructed to carry out the mechanical blade-cleaning step wherein the bottom of the shear blade was brushed. The brush assembly was removed from the sample collection station and set aside for weighing at a later time.
- (6) Finally, the computer program proceeded to the next segment, as appropriate, by repeating the shearing cycle, or indicated to the operators that the rod shearing was complete and that they should proceed to the next operation, namely, weighing of the sheared segments.

7. Weighing the Sheared Segments

When shearing of the fuel rod was complete, the capped aluminum cans that contained the sheared segments were individually weighed. Each full can was weighed on one or the other of two in-cell balances. Full cans weighing less than 300 g (net segment weight less than 286 g) were weighed to 0.001 g with the remotely operated 300-g capacity balance; those weighing 300 g or more were weighed to 0.01 g with the remotely operated 2000-g capacity unit. The operators were prompted through the weighing operations by the computer, which selected the appropriate balance for each segment by referring to the estimated segment weights coded in the FRIF. Each full can weight was determined as the average of three separate weighings (can weight minus average balance tare) of the can. Each individual weighing result for each segment was recorded in the FSS process log and in the summary data file for the fuel rod.

8. Weighing of Material on Brushes

Material collected on the brushes used for intersegment cleaning of the shear blade included both fine particles of ceramic fuel and Zircaloy cladding fines. In general, the Zircaloy fines were large enough to be visually identified and removed from the brush surface with forceps. Because of this

characteristic of the fines, the weights of both materials on each brush could be separately determined in the computerized procedure for the post-shearing brush weighing.

In this procedure, the operator was prompted to weigh each brush assembly to 0.001 g, and the computer calculated the total weight of material on the brush by using this weight and the pre-shearing brush weight determined previously. Then, the operator was prompted to remove any Zircaloy fines that might be present on the brush and to reweigh the assembly. Using this brush weight and the first post-shearing brush weight, the computer calculated the apparent weight of the metal fines that were removed. Both the total weight of material on the brush and the weight of the Zircaloy fines were recorded in the FSS computerized log and in the summary data file for use in subsequent material balance calculations.

9. Estimation of Fission Gas Released during Shearing of Each Segment

The weight of fission gas released from the ceramic fuel during shearing of each segment was estimated from ^{85}Kr release data obtained by integrating the output from the shear-cell stack monitor over the time period in which the segment was sheared. In relating the measured ^{85}Kr release to the total weight of released fission gas (i.e., Kr plus Xe), we assumed that the composition and isotopy of the fission gas freed by shearing was the same as the composition and isotopy of the fission gas recovered from the rod plenum.

During the shearing operations, the integrating devices on the stack monitor system were reset each time shearing of a segment was begun, and the accumulated integral was manually recorded when the shearing of the segment was complete. The stack-monitor output was also monitored continuously during the shearing by means of a strip-chart recorder. Individual signal spikes, each corresponding to a burst of gas containing 10 to 200 μCi ^{85}Kr being released during the shear stroke through the segment, were readily apparent in the recorder tracing (see example in Fig. 13), giving an indication of the sensitivity of this monitoring system.

Compilation, reduction, and reporting of the data on gas release during shearing were performed manually, rather than by computer, because some aspects of the data interpretation (e.g., selecting baselines, correcting for gas distribution between the two exhaust stacks in the cell) required subjective judgments to be made. Since the measurement of fission gas released during shearing was not a requirement but was requested by BAPL to be carried out on a best-effort basis, little effort was expended toward streamlining and fine-tuning these operations. Of course, all reasonable care was taken during the collection and processing of the data to provide the best practical measurement with the equipment that was available. Additional information regarding the determination of the gas release during shearing is contained in Section V.G, wherein results of the measurements are discussed.

10. Material Balance Calculations and Data Summary

When all of the FSSF operations described above were completed for a given fuel rod, the data were processed, with a stand-alone report-generation program, which generated a shearing data summary as illustrated in Fig. 14. This program read the rod's FRIF and the summary data file generated during

SEGMENT

Fig. 13.

Example of Strip-chart Recorder
Tracing for Fission Gas Release
during Shearing

FSSF operations with the rod, calculated results for the various measurements performed, and wrote the results to a new file in a format appropriate for management review or transmittal as a report on the shearing of the rod. As Fig. 14 shows, the summary has three parts. The first part identifies the rod and gives results of measurements made on the rod; the second part gives a tabular summary of information related to individual segments sheared from the rod; the third part presents a graphical representation of the shearing plan for the rod and compares the desired and actual locations of each segment boundary.

FULL SCALE SHEAR
FUEL ROD SHEARING DATA SUMMARY
20-AUG-84

FUEL ROD SERIAL NUMBER	26-06481
ANL IDENTIFICATION	B
ROD DIAMETER (in.)	0.527
FUEL ROD LENGTH (in.)	118.191
FUEL ROD TEMPERATURE AVG. (C)	33.05
FUEL ROD WEIGHT BEFORE PUNCTURE (g)	3555.29
FUEL ROD WEIGHT AFTER PUNCTURE (g)	3555.40
TOTAL ROD RECOVERY (g)	3552.567
TOTAL ROD RECOVERY (%)	99.920

SEGMENT DATA SUMMARY

SEGMENT	START POSITION (in.)	END POSITION (in.)	SEGMENT LENGTH (in.)	SEGMENT WEIGHT (g)	BRUSH RECOVERY, (g)		TOTAL RECOVERY (g)	LINEAR DENSITY (g/in.)	TOTAL SHEAR CUTS
					FUEL	ZIRCALOY			
00	118.191	107.199	10.993	186.831	0.000	0.000	186.831	16.996	29
01	107.199	96.207	10.992	344.533	0.063	0.000	344.596	31.350	44
02	96.207	78.116	18.091	571.146	0.077	0.000	571.223	31.576	73
03	78.116	60.616	17.500	552.120	0.073	0.000	552.193	31.554	70
04	60.616	43.116	17.500	551.396	0.045	0.000	551.441	31.511	70
05	43.116	25.619	17.497	550.561	0.041	0.003	550.605	31.468	70
06	25.619	11.057	14.562	460.306	0.058	0.000	460.364	31.615	59
07	11.057	1.625	9.433	299.025	0.024	0.000	299.049	31.703	36
08	1.625	0.000	1.625	36.258	0.008	0.000	36.266	22.322	4
TOTALS			118.191	3552.175	0.396	0.003	3552.567		455

FUEL ROD SEGMENT LOCATIONS, REQUESTED ALL DIMENSIONS IN INCHES
AS-CUT

was determined and might not be representative of the overall average surface temperature of the rod.

The fuel rod weights before and after puncture of the rod plenum are those determined as described in Section IV.D.4. Any apparent difference between the two weights should be attributed to imprecision of the weighings rather than to an actual change in rod weight. Calculations to predict the weight change associated with removal of the plenum gases and filling of the void volume with air indicate that it would be only a few milligrams and would not be detectable with the balance used in weighing the rod.

The total rod recovery, both in terms of weight and percent of rod weight, was computed as the sum of the weights of all rod components that could be accounted for after the rod was sheared. Rod recovery includes material recovered on the brushes used in cleaning the shear blade, as well as material collected in each aluminum sample can. This recovery value should not be confused with the fuel recovery fraction, which is computed from the weight of fuel lost during shearing (which can be estimated from the ANL data) and the fuel stack weight for the rod of interest; this latter information was known only to BAPL. The total rod recovery values listed in the data summary provide a useful gauge of material balance during shearing of the rod and a working standard for evaluating ANL's performance in shearing each rod; unaccounted material was assumed to be fuel, rather than cladding plus fuel. Obviously, high total rod recovery is a prerequisite to satisfactory fuel recovery during shearing.

In the Segment Data Summary, the start and end positions for each segment correspond to the measured, actual boundary cut locations at the two segment ends as described in Section IV.D.6; the segment length was computed as the difference between these positions. The segment weight was calculated as the difference between the full can weight (Section IV.D.7.) and the corresponding empty can weight (Section IV.D.1.c.). The weights of fuel and Zircaloy recovered on each brush assembly were obtained from the post-shearing brush weights as described in Section IV.D.8. Total recovery for each segment was computed as the sum of the segment weight and the brush recovery results. The linear density of the segment was simply calculated as the ratio of the total recovered segment weight to the segment length.

The graphical representation of segment location in Fig. 14 should be self-explanatory.

11. Post-Shearing Operations

When shearing operations (including the weighing of segments and brushes) were complete and the shearing data summary had been reviewed by the shear operators, the segments from the rod were transferred to the DDS facility by means of the can-out transfer system, and the equipment in the FSSF was prepared for operations with the next fuel rod. All segments were transferred to the DDS, except for the stub segment, which was not intended for dissolution. Periodically, when several cans containing the stub ends had accumulated, the cans were opened, the contents were examined and weighed to verify that each stub was completely recovered and that fuel particles were absent, and then the materials were discarded to waste.

Before shearing the next rod was started, the shear tooling and the sample collection station were removed from the shear base, were disassembled, and were cleaned with Freon solvent to remove any fuel material that might have adhered to the surfaces of the components. Progress in the cleaning operations was monitored by measuring the gamma radiation emitted by the component being cleaned. An in-cell portable radiation meter was used for this measurement. In general, cleaning of the shear tooling resulted in a decrease in activity from about 5 R/h at two inches to only a few hundred mR/h at two inches. Quantities of fuel corresponding to these latter levels of activity were too small to be visible and were judged to pose no threat of contamination due to carryover into segments sheared from subsequent rods.

E. Operation of the Dual Dissolver System (DDS)

The primary objective of operations with the DDS was to individually dissolve each sheared segment and thereby put the segment fuel material into a homogeneous form (solution) that could be conveniently and representatively sampled for analysis. Operations were carefully managed to serve specific analytical requirements of the dissolution process and to promote efficient use of equipment and manpower during the three-shift operation schedule (five days per week, 24 hours per day).

The DDS operations employed a two-stage dissolution process, wherein the segment was twice contacted with dissolvent, and the resulting solutions and dissolver rinses were transferred into a blend tank (BT) where they were thoroughly mixed before being sampled. Provision was made in the procedures to (1) collect and sample fission gases (Xe and Kr) released during dissolution of the segment, (2) assess the completeness of the segment dissolution as well as analytically characterize the solution in the BT, and (3) carry out an in situ spiking of the BT solution as part of the mass spectrometric isotope dilution (MSID) assay of uranium in the segment. The entire sequence of operations was carried out by means of the computer-directed operating procedure, OPRDSx.

In the following subsections, we describe the various aspects of the DDS operations, beginning with the management considerations that were applied in planning the processing of segments from each individual LWBR fuel rod, proceeding to the architecture of the OPRDSx computerized procedure, and ending with a description of major operations within the procedure. As was the case with the FSSF operations, the descriptions are not intended to be exhaustive; any reader interested in the step-by-step execution of the DDS procedure, which involved more than 350 separate steps for processing an individual segment, should refer directly to the procedure documentation.⁹ Equipment in the DDS facility, which is referred to in the following discussions, is described in Section III.A.4.

1. Management Considerations

Specific guidelines regarding management of the dissolver operations were established for the activities that preceded dissolution of the group of segments from each individual fuel rod. (Segments from only one rod at a time were permitted in the DDS hot cell.) These guidelines provided a standardized approach to (1) the assignment of a dissolution order for segments from the rod, (2) cleanup of a dissolver after all the segments from a given rod were processed, and (3) designation of equipment items to be used for the individual segment

dissolutions. This process-planning information was entered, by the DDS task leader, into the FRIF, for subsequent retrieval by the computerized procedure during dissolver operations (see Fig. 9 for an example of the FRIF for an individual rod).

A computerized procedure was developed to assign the individual segments to one or the other of the two dissolver systems, to establish the order for dissolution of segments assigned to each dissolver, and to minimize the potential impact of residual traces of fuel from one segment being carried over in the dissolver to the next segment dissolution. In this system, the segments from the rod of interest were first divided into two categories: a "low-uranium" category and a "high-uranium" category, based on the approximate segment uranium loadings as provided by BAPL in the shear plan for the rod. The low-uranium segments were usually dissolved in DS1, and the high-uranium segments in DS2. Within each category, the segments were generally dissolved in order of increasing uranium content.

Following dissolution of the final segment from a particular fuel rod in each dissolver, a "blank" or "cleanup" dissolution (containing no segment) was performed. The blank dissolutions were assigned dummy segment numbers between 91 and 99 in the FRIF. Odd numbers indicated a blank run in DS1, and even numbers indicated a blank run in DS2.

These blank dissolutions served two purposes. First, they reduced the quantity of fuel residue that might be carried over into the first segment of the next rod to be processed in the dissolver. Second, the uranium recovered in the blank run provided a means to estimate the amount of fuel carryover that might have taken place between dissolutions. In some cases (e.g., just prior to dissolving segments from the reflector rods, which had very low uranium loading compared to the rods processed immediately before them), two blank runs were scheduled for each dissolver to improve the dissolver cleanup.

The equipment items specified in the FRIF were those reusable components (such as BTs, filter assemblies, and gas-sampling cylinders) that required decontamination, replacement of individual disposable parts, or some other treatment before being considered acceptable for reuse. Whenever practical, e.g., in the case of filter assemblies, reuse of a given item was scheduled for every other rod so that one group of items could be cleaned, assembled, and tested during the time period when another group was being used. This approach helped maintain continuity in the operations and minimized dissolver downtime caused by equipment not being available when needed.

2. General Features of the Computerized DDS Operating Procedure

Segment dissolutions in the DDS followed an operating procedure that was originally developed for GRIP-II fuel material³ and later expanded and computerized prior to the EOL campaign. As with the computerized procedure used for FSSF operations (OPRFSS, Section IV.D), the computerized dissolution procedure used the tools provided by the POBSYS software system to provide step-by-step instructions for the operator by means of terminal display, to carry out process-monitoring functions and fully or partially automated process-control operations, and to acquire and record data.

Each of the two dissolver systems was run from its own version of the computerized procedure (designated OPRDS1 and OPRDS2 for dissolver systems DS1 and DS2, respectively, or OPRDSx for the general case). The two procedure versions were identical, except that addresses of equipment interfaces and peripherals (such as terminals and printers) were specified to correspond to the components in the appropriate dissolver system. This arrangement permitted the two dissolvers to operate simultaneously and independently of each other. The OPRDSx procedure had many of the features described for OPRFSS, including continuous monitoring of sensors and display of alarms, specification of information from the FRIF or other files to aid the operator in executing the operations, and automatic logging of process information, both in computer files and hardcopy.

The OPRDSx procedure also had features designed to accommodate managerial and operational requirements peculiar to the dissolver operations. Particularly notable among these features were (1) a means for maintaining an up-to-date inventory of reusable equipment using the files associated with the HISLOG program for tracking equipment usage [see Section III.C.2.c.(3)]; (2) an interactive process-control (IPC) program which permitted an operator to adjust process conditions through computer command without having to exit the main program and thereby interrupt ongoing monitoring or automated activity; and (3) graphical displays, including multicolor dynamic piping and instrumentation (P&I) diagrams (i.e., schematic illustrations of system components that were continuously updated in real time by the computer to show the current on/off, open/closed, etc., status of each component), and plots of selected temperature or pressure data vs. time during specific operations.

Whenever a listed reusable item was called for in executing a procedure, the OPRDSx programs checked the HISLOG file appropriate to each equipment item specified in the FRIF (or by the operator) to verify that the item was ready for reuse. Items that were not available were rejected by the program; action was then required of the operator (or shift supervisor) to rectify the situation. Also, during execution of the procedure, the program automatically updated the HISLOG files to indicate the segment number for which the item was used, and it noted in the files the completion of operations involved in preparing the item for reuse, including any pertinent data obtained during these operations. Examples of HISLOG files for a typical BT, gas-sampling cylinder, and filter assembly are shown in Fig. 15.

The IPC programs were an adjunct to the automated process-control system used by OPRDSx to carry out operations with DS1 or DS2. With these programs, the operator could select and execute, by a terminal command, any of the 23 process-control states available to the automated system. Each process-control state is associated with a particular function (or operation) and corresponds to a specific configuration of valves, pumps, etc., within a given dissolver system. A listing of the process-control states for one (DS2) of the two dissolver systems is shown in Table 6. The corresponding states for DS1 have identical identifications (e.g., PCS-01) but involve valves and pumps that have different (odd) numbers.

These IPC programs provided a means for dissolver operators to respond to alarm conditions, troubleshoot system leaks identified during pre-dissolution testing, and work around minor equipment problems that might otherwise have aborted the operating programs and caused lengthy, unnecessary delays in completing the segment dissolutions. These programs contained

BLEND TANK : TK-03

ENTRIES BY SHIFT SUPERVISOR ONLY

DISS. RUN	DISS. START	INITIAL WT, g	DECONTAMINATED WT, g	BY	DATE	BT SAMPLE TO ACL	BY	U ANALYSIS ug	BY	DATE	REUSABLE Y/N	BY
B-03	1082084	7284	7283	HM	090784	091184	JH	17	LC	091384	Y	FM
C-03	1091984	7283	7283	DR	100884	092884	TC	413	LC	100484	N	FM
D-03	1091984	7283	7283	DR	100884	101084	TC	11	LC	101684	Y	FM
D-03	1102184	7283	7283	FM	102684	102984	FM	153	LC	111084	N	FM
D-03	1102184	7283	7284	JK	110984	110984	DR	7	LC	111584	Y	FM
E-05	1111984	7284	7283	FM	112184	112684	TC	10	LC	113084	Y	FM
F-04	1120484	7283	7283	LR	120884	121184	RN	16	LC	121884	Y	JH
G-06	1011485	7286	7282	CW	011785	011785	TC	16	LC	012385	Y	TC
H-15	1012485	7283	7283	TC	012585	012585	DR	5	LC	020185	Y	DG
H-04	1020585	7284	7282	HM	021285	021385	DR	4	LC	021985	Y	FM
I-08	1022085	7283	7283	HM	022285	022285	JH	5	LC	030485	Y	TC
J-02	1031185	7282	7283	HM	031485	032185	DR	6	LC	032785	Y	FM
K-03	1041585	7289	7282	DP	041985	042285	DR	16	LC	050285	Y	FM
L-02	1051385	7283	7282	JK	052385	052385	DR	19	LC	052985	Y	FM
W-01	1060385	7283	7282	HM	060585	060585	DR	21	LC	061385	Y	TC
M-03	1061785	7284	7282	HM	062085	062185	TC	6	LC	070885	Y	TC
N-03	1070885	7283	7282	WK	072285	072285	DR	23	LC	072685	Y	DR
O-04	1073085	7282	7281	LR	080685	080685	TC	11	LC	081385	Y	FM
P-92	1090285	7283	7282	FM	090385			0			Y	FM
Q-02	1090385	7284	7281	DP	091685	091685	DR	5	LC	092585	Y	FM
R-08	1100185	7283	7282	DP	100485	100485	FM	3	LC	101185	Y	TC
R-07	1101385	7282	7281	WK	101785			0			N	FM

GAS SAMPLING CYLINDER: SC-02

DISS. RUN	DISS. START	CYLINDER S. SC-	GAS MASS SPEC TO	BY	FROM	BY	CYLINDER PURGE SC-	DATE	BY	REUSABLE Y/N	DATE	BY
B-02	1082084	02 12	082284	DP			02	090684	FM	Y	090684	FM
C-03	1091984	02 12	092084	DR	092784	DR	02	102684	LR	Y	102684	LR
D-05	1102984	02 12	103084	DR	110284	FM	02	110684	DR	Y	110684	DR
F-02	112684	02 12	113084	TC	120584	TC	02	120684	DP	Y	120684	DP
G-02	121184	02 01	121284	DR	121884	DR	02	121884	FM	Y	121884	FM
H-05	1012185	02 01	012285	TC	012485	TC	02	020185	FM	Y	020185	FM
H-03	1020385	02 01	020585	DR	021285	DR	02	021585	CW	Y	021585	CW
I-04	1021885	02 01	021985	TC	022185	TC	02	022285	LR	Y	022285	LR
J-01	1031285	02 01	031385	TC	031985	DR	02	032085	LR	Y	032085	LR
K-02	1040985	02 12	041185	WK	041685	DR	02	041985	DP	Y	041985	DP
L-02	1051385	02 12	051585	WK	052385	WK	02	052985	WK	Y	052985	WK
W-02	1052985	02 12								Y	061485	FM
M-02	1062585	02 12	062085	DP	070985	TC	02	071685	TC	Y	071685	TC
N-02	1071685	02 12	071785	TC	072485	TC	02	072485	TC	Y	072485	TC
O-02	1080185	02 12	080285	TC	080685	TC	02	080785	JK	Y	080785	JK
P-02	1082785	02 12	082985	CW	090485	FM	02	091185	DP	Y	091185	DP

SOLUTION FILTER ASSEMBLY: FR-01

DISS. RUN	DISS. START	PAPER REMOVED DATE BY	DECON. DATE BY	ASSEMBLY DATE BY	LEAK CHECK DATE BY	REUSABLE Y/N	BY
B-01	1082184	109184 FM	8	1092684 FM	1101684 FM	Y	FM
D-01	1102184	102584 FM	8	1102584 FM	1102584 FM	Y	FM
F-00	112684	121284 FM	6	1121284 FM	1121384 TC	Y	FM
G-06	1011485	1012185 DP	6	1012285 FM	1012285 FM	Y	FM
H-06	1012385	1021485 FM	15	1021485 FM	1021585 TC	Y	TC
I-00	1021885	1022285 TC	4	1022285 TC	1022585 FM	Y	FM
J-00	1030585	1032785 TC		1032785 TC	1032885 TC	Y	TC
K-01	1040285	1040485 TC	7	1040485 TC	1040585 JK	Y	JK
W-01	1060385	1060485 TC	7	1060485 TC	1060585 FM	Y	FM
N-01	1071585	1072385 WK	6	1072385 WK	1072385 DP	Y	DP
O-01	1073085	1080585 WK	8	1080585 WK	1080585 EK	Y	WK
Q-01	1090985	1091185 FM	6	1091185 FM	1091185 FM	Y	FM
R-01	1100285	1100485 FM	4	1100485 FM	1100485 FM	N	FM

Fig. 15. Examples of HISLOG Files

Table 6. Dissolver 2 Process Control States

Process-Control State No.	Function	Components Operated ^a
PCS-00	System Deenergized - Reset or Initial State	All valves closed, pumps off
PCS-01	Charge Reagent to Dissolver through the Spray Ring	CV2, CV4, CV6, CV18, CV20
PCS-02	Charge Reagent to Dissolver through Drain Line, Backflushing Solution Filter	CV2, CV8, CV10, CV18, CV20
PCS-03	Charge Reagent to Blend Tank	CV2, CV12
PCS-04	Transfer Solution from Dissolver to Blend Tank	CV8, CV10, CV12, CV18, CV20, CV34
PCS-05	Sample Dissolver Solution	CV8, CV10, CV14, CV18, CV20
PCS-06	Pressurize Dissolver with Dry Air to 120 psig	CV18, SV44
PCS-07	Pressurize Dissolver with Dry Air to 70 psig	CV18, SV46
PCS-08	Vent Dissolver to Hold Tank	CV16, CV22
PCS-09	Pump Dissolver Head Space Gas to Hold Tank	CV16, CV24, P4
PCS-10	Vent Hold Tank to Cell (Rapid Vent)	CV24, CV30, CV32
PCS-11	Vent Hold Tank to Cell (Controlled Release)	CV24, CV32
PCS-12	Recirculate Hold Tank Gas, Without Sampling	CV22, CV24, P4
PCS-13	Recirculate Hold Tank Gas, With Sampling	CV22, CV26, CV28, P4
PCS-14	Pump Down Hold Tank, Vent to Cell Exhaust	CV22, CV30, CV32, P4
PCS-15	Flush Off-Gas System with 70 psig Dry Air	CV16, CV18, CV22, CV26, CV28, CV30, CV32, SV46
PCS-16	Initiate Full Condenser Cooling Water Flow	SV42
PCS-17	Cool Dissolver Vessel	SV42, BL2
PCS-18	Evacuate Blend Tank Head-Space Gas (Controlled Venting)	CV36
PCS-19	Evacuate Vacuum Reservoir Tank	P10
PCS-20	Evacuate Blend Tank and Vacuum Reservoir Tank	CV34, P10
PCS-21	Evacuate Solution Filter	CV10, CV12
PCS-22	Air Purge of Gas Sampling Cylinder	CV22, CV28, CV30, CV32, P4

^aComponent designations are CV = Control Valve, SV = Shutoff Valve, BL = Blower, and P = Pump.

built-in software interlocks to prevent the operator from activating certain critical process-control states under conditions that could jeopardize the validity of analytical results obtained for the segment being dissolved. In a few cases, the need for additional interlocks during specific operations was not recognized until after operational mishaps had occurred. (See Section V.C.3.b for a discussion of operational mishaps during the dissolutions of LWBR fuel rod segments and successful recovery actions that were taken to deal with them.) The IPC programs were subsequently modified as appropriate during the EOL campaign to guard against recurrence of each type of problem that arose.

Owing to the graphical displays in the computerized dissolver operations, the operator could examine the status of system components or monitor the progress of a particular operation (e.g., heatup or cool down of a dissolver vessel or BT) in a straightforward, easy-to-read format. Examples of the P&I diagrams and plots that were available for display are shown in Fig. 16. Appropriate displays were called up by the OPRDSx programs during certain automated procedure steps; any of the displays could also be called up by operator command at the work station terminal, as desired. Data used in the plotting displays were stored in special computer files regardless of whether or not the display was active at the time the operations portrayed in the plot were carried out. The data in these files could be plotted at any time, on command, so that the dissolver operator could switch between displays without losing information, or the operations could be reviewed after they were complete.

Data acquired by the computer system during each segment dissolution, including operator sign-off information, alarm messages, and operator comments, were recorded in a manner analogous to that used at the FSSF. Detailed information was recorded in a computerized process log (both computer file and printed versions), and selected information was stored in a computerized data summary file for subsequent use in generating reports. Several manual logbooks were also maintained at the DDS work stations. A manual operations logbook was kept for each dissolver station, in which operator comments and actions, important data, unusual occurrences, etc., were noted on a segment basis. A separate, general operations log was also kept at the work area to note activities related to the entire DDS facility. Specific routine activities were documented in supplemental logbooks titled according to the activity, e.g., Equipment Maintenance, Sample Movement, and Fissile Material Inventory. Complete, redundant records, as provided by this data management system, formed the foundation for quality assurance within the project.

The OPRDSx procedure for dissolving segments in the DDS dissolver systems recognized three types of segment on the basis of the segment identification number: (1) a fuel-bearing segment, identification number in the range 01 to 17; (2) a plenum segment, number 00; and (3) a dummy segment, number in the range 91 to 99. Details of the dissolution sequence were automatically varied by the procedure programs to correspond to the type of segment being dissolved.

3. The DDS Operating Procedure

The DDS operating procedure had five major sections: (1) preliminary equipment checks and installations of equipment items, (2) the two-stage dissolution of the segment, (3) sampling of the dissolver off-gas, (4) BT

Fig. 16. Examples of Dissolver System Graphics

operations, and (5) post-dissolution operations, such as recovery of the segment's cladding hulls and decontamination of reusable equipment. All of these operations were carried out for each fuel-bearing segment. For plenum and dummy segments (i.e., cleanup dissolutions), certain operations were automatically omitted or modified by OPRDSx to streamline the process; these exceptions are identified in the appropriate subsections. A complete dissolution for a fuel-bearing rod section required 30 to 48 h.

a. Preliminary Operations

In preparation for a given dissolution, operations were carried out to check performance of the equipment and to provide the needed materials; these operations are discussed below.

(1) Balance Tests

The in-cell balances to be used during dissolution of a given segment were first calibrated with the in-cell standard weights and then individually tested for linearity, precision, and stability by means of the BALQA balance-test program [Section III.C.2.c.(1)]. Each time one of these balances was specified for use during subsequent operations, OPRDSx checked the computerized record of tests performed on the balance and determined the time since the last successful BALQA test was run. If the prescribed balance had not passed BALQA within the previous 48 h, OPRDSx automatically brought up the BALQA program for the operator to execute before proceeding with the weighing operations.

In a similar fashion, the short calibration-check program, BALCC, was brought up by OPRDSx just prior to each weighing operation in the procedure, even if the requirement on BALQA was satisfied. If the balance failed the BALCC check, BALQA was, again, brought up automatically for the operator to run as an aid in diagnosing the reason for the failure. These precautions helped assure the validity of weighing data obtained during each DDS operation.

Although the same balance-testing procedures were used in the FSSF and DDS (cf. Section IV.D.1.b. for the FSSF operations), the timing of the tests and the tolerances applied were different for the two facilities so that the specific analytical and operational needs of the respective use of the balances could be accommodated.

(2) Weighing of Sample Bottles

Another preliminary operation to each segment dissolution was the weighing, both outside the cell and inside, of individual empty glass bottles, used for sampling the dissolver solutions. This was done to establish a tare value for each sample taken. Prior to weighing, each individual bottle and its cap were labeled with the identification code of the sample for which it was to be used; the bottle and cap were weighed together in determining the empty-bottle weight.

During the weighing routines, the computer prompted the operator to weigh, in triplicate, to the nearest milligram, the appropriate number of bottles for the type of segment dissolution being performed, i.e., ten bottles for a fuel-bearing segment, four bottles for a plenum segment, or two bottles for a blank run. The computer called for each bottle on the basis of the identification code on its label; such specific prompting by the computer was made possible by the standardized sample nomenclature described in Section IV.A.

The OPRDSx routine for weighing the bottles inside the cell also compared the in-cell weight of each bottle with the out-of-cell weight and advised the operator concerning the significance of any differences that were

detected. This comparison served to check the performance of the in-cell balance, as well as to reduce the chance that the sample bottles had been mixed up during one or the other weighing. The out-of-cell bottle weights were recorded in a special computer file; the in-cell weights and the differences between the in-cell and out-of-cell weights were recorded in the dissolution process log and the dissolver data summary file.

(3) Installation of Filter Assembly, Gas-Sampling Cylinders, and BT

For each segment dissolution, we installed fresh auxiliary components in the dissolver system, including the filter assembly in the solution transfer line, gas cylinders used to obtain samples of the dissolver off-gas, and a BT. Before this installation, the OPRDSx program instructed the operator to select the component predesignated in the FRIF for use with the given segment and, by reading the HISLOG file, it verified that the component had been appropriately decontaminated and tested for reuse since its last previous use in the system. The decontamination procedures for the filter assemblies and BTs are described in Section IV.E.3.f.(1) and (2). If any of the designated components were not verified as ready-to-use, the operator was able to make a suitable substitution; the substitute component was also checked by the program to verify its acceptability. When the components were accepted, their identities were automatically recorded in the process log and data summary for the segment being processed. The operator was then instructed to install the components.

Installation of the BT included weighing of the BT support assembly, both with and without the tank in place; each weighing was made in triplicate. The weight of the complete assembly was stored in the computer for use in computing net weights of solution in the BT during subsequent operations; the difference between the weights with and without the BT was taken as the weight of the empty BT itself. The empty BT weight was recorded in the process log, the dissolution data summary, and the HISLOG file for that BT. The HISLOG record of empty weights for each of the nine reusable BTs served as a means of monitoring corrosion of the tanks with use; weight changes for the BTs proved insignificant over the duration of the EOL campaign, suggesting minimal corrosion.

Gas-sampling cylinders were required only for dissolutions involving fuel-bearing segments. In the case of plenum or blank dissolutions, where off-gas was not sampled, the operator was instructed to install a jumper connector in the off-gas sampling line in place of the gas-sampling cylinders.

(4) Loading of the Segment into the Dissolver Vessel

After the installations of the components, the operator was prompted by OPRDSx through a check of the helium and air supply systems associated with the dissolver system and then was instructed to load the segment (in its capped aluminum can) into the dissolver vessel. The operator first retrieved the specified segment from the shielded segment-storage area and weighed the segment on the appropriate (i.e., 300-g or 2000-g capacity) balance. The computer compared the result of this weighing with the appropriate full-can weight logged during FSSF operations as a means to verify the identity of the segment. If the DDS and FSSF weights did not agree within predetermined limits (a situation that did not arise during actual EOL operations), the operator

was instructed to double-check the identity of the selected segment and to enter a comment to the computerized log before proceeding. As an additional quality assurance (QA) check to verify the identity of the segment before it was dissolved, the operator was prompted to have the shift supervisor enter the identity of the segment being prepared for dissolution. If the supervisor's entry did not agree with the information previously entered to OPRDSx, the program indicated the discrepancy and required an additional entry by the supervisor, acknowledging that the discrepancy had been resolved. Once the problem had been solved, operations could proceed. The segment identity, the FSSF and DDS segment weights, and the weight difference were automatically written to the process log and the dissolver data summary file.

Before the segment was placed in the dissolver, the cap of the aluminum sample can was punctured with a specially built device. The purpose was to equalize the pressure when the dissolver vessel was pressurized and depressurized during pre-dissolution operations. This prevented the can from being deformed and allowed its retrieval, intact, if some mishap that required drastic action occurred during these preliminary operations.

To load the segment, we removed the dissolver cover flange and installed a new flange gasket. The segment was then locked into the tantalum dissolver basket and lowered into the vessel. The vessel was sealed by bolting the cover flange in place.

(5) Pre-Dissolution Leak Testing

Once the segment was loaded into the dissolver vessel and the vessel was sealed, the computer carried out a series of fully automated leak tests on the dissolver vessel, the BT assembly, the filter assembly, and the off-gas system. In each test, an appropriate portion of the dissolver system was either pressurized or evacuated, and the pressure was monitored at specified intervals (usually 5 to 10 seconds) over a preset period of time. If the pressure changed by more than a prescribed tolerance during the test, or if the prescribed starting pressure for the test could not be attained, the computer notified the operator that a leak was present and that the leak should be repaired and the test repeated.

The dissolver vessel was pressurized with air to 120 psig for the test and was required to lose no more than 2 psig in 15 min to pass the leak test. The air from the dissolver vessel was then transferred to the off-gas system and used to pressure-test it. The off-gas system was required to lose no more than 1 psig pressure, from a starting pressure of not less than 10 psig, during a five-minute recirculation of the gas in the system. The BT and filter assemblies were tested under static vacuum at -9 psig or less. The BT assembly was judged acceptable if its pressure did not increase by more than 0.5 psig in five minutes. The filter-assembly pressure had to increase by not more than 0.5 psig over a two-minute period to be judged satisfactory. When all the components had successfully passed their leak tests, dissolution of the segment was begun.

b. Two-Stage Segment Dissolution

Dissolution of each fuel-bearing segment was a two-stage process in which the segment was subjected to a primary and then a secondary contacting with dissolvent at elevated pressure and temperature. After each contacting, the solution in the dissolver was transferred into the BT and the dissolver vessel was given a special rinse treatment; each rinse solution was also transferred to the BT. Samples of the solution from the secondary contacting were taken, during transfer of the solution to the BT, to assess the completeness of segment dissolution. Gases collected in the dissolver off-gas system during the primary contacting were also sampled to determine the fission gases that were released by dissolving the segment.

In carrying out these operations, the OPRDSx computer programs alternated between interactive sequences, in which the DDS operators were instructed to perform specific manual manipulations, and fully automated sequences wherein the computer (1) monitored key sensors to evaluate the status of operations and initiated progressive stages of the process, (2) performed process-control activities to configure valves, etc., in a manner appropriate to the operations at hand, and (3) calculated heat and material balances to aid the operators in tracking progress of the operations and performance of the equipment. Operating conditions and sequences for the primary and secondary dissolutions are described below; sampling of the dissolver off-gas is discussed in Section IV.E.3.c.

Three different reagents were used in the DDS operations: Thorex reagent (13.6M HNO_3 , 0.06M HF with Cs and Ce carriers at 1 $\mu\text{g/mL}$ concentration); Thorex-Al reagent (identical to Thorex, but containing 0.06M Al^{3+}); and dilute (0.5M) nitric acid used as a rinse solution. The Cs and Ce carriers in the Thorex reagents prevented adsorptive losses of these fission-product elements from the fuel solution. The Al^{3+} in the Thorex-Al solution complexed the fluoride ion during the secondary dissolution (for which the Thorex-Al reagent was used), and thereby inhibited corrosion of the tantalum vessel. The aluminum from the segment sample can served the same purpose in the primary dissolution, where the standard Thorex reagent was employed.

Each reagent was prepared in 50-L batches by the ACL and was introduced to the DDS through the reagent-charging station. Each batch was analyzed, at the time it was prepared, to verify its composition and to demonstrate the absence of significant uranium impurities. Details of the preparations and analyses are given in Document C-0030-0286 (see Attachment 1). The ACL issued a certificate of analysis for each reagent batch, which listed the results of the composition verification, as well as other information pertinent to the traceability of the data (see Attachment 2 for examples of these certificates).

(1) Primary Dissolution

For all primary dissolutions, the segment was contacted with Thorex reagent at 195°C under 120 psig pressure for four hours; for blank dissolution runs, the primary dissolution was omitted completely. The volume of Thorex used for any given segment was specified in the FRIF and was based on segment type, weight of fuel material, and experience gained during prior

dissolutions of similar segments. Thorex volumes ranged from 1.0 to 3.5 L for the LWBR fuel rod segments. Upon completion of the primary dissolution, the solution was transferred to the BT, and the dissolver vessel was rinsed by refluxing 750 mL of dilute nitric acid for 15 min at 100°C under 10 psig pressure. This reflux-rinse solution was also transferred to the BT.

In carrying out the primary dissolution, the OPRDSx program first referred to the FRIF to identify the Thorex volume specified for the segment being dissolved and then instructed the operator to dispense the appropriate weight (computed using the Thorex density) of Thorex reagent into the charging vessel at the reagent-charging station. After the reagent dispensing was complete, the operator was requested to enter the type of reagent (entry checked to verify "Thorex"), the batch number of the Thorex used, and the weight of reagent dispensed. Each entry was recorded in the computerized log and data summary file.

Upon accepting the reagent identity and quantity, the computer (1) automatically checked specific sensors to verify that the dissolver was in proper condition to receive reagent, (2) energized the appropriate process-control state to transfer reagent into the dissolver vessel through the spray ring, (3) displayed the appropriate P&I diagram on the graphics terminal, and (4) pumped the Thorex from the charging station to the dissolver vessel. It then checked the dome-valve control pressure and air supply pressures, pressurized the dissolver with 70 psig air, and initiated full cooling water flow to the condenser. When satisfactory execution of each operation was verified, the operator was instructed to turn on the heater controls and reset the kilowatt-hour (kW-h) meter for the dissolver system. When the operator acknowledged completion of these steps, the program began a fully automated heatup procedure.

For the heatup sequence, the program preset alarm criteria for specific sensors in the system, displayed a real-time plot of temperature vs. time on the graphics terminal, and then energized the lower-zone (LZ) heaters on the dissolver, monitoring the kW-h meter to determine whether all heaters were functioning and whether adequate heating was available. Abnormally low or inadequate heat flow to the dissolver was called to the operator's attention and corrective action (e.g., stopping the heatup to allow for operator review) was automatically taken as required. Once proper operation of the LZ heaters was verified, the program checked the FRIF to determine whether the upper-zone (UZ) heaters were required for this dissolution. (The UZ heaters were used for Thorex volumes of 3.0 L or greater.) If needed, the UZ heaters were automatically started in a manner analogous to that used for the LZ heaters. The program then entered a sensor monitoring-and-display sequence to track progress of the heatup. When the LZ heater temperature reached a value of 150°C, the operator was instructed to retighten the bolts on the dissolver cover flange; this step in the procedure was added because small vapor leaks were occasionally found at the closure gasket during heatup with the early LWBR fuel rod segments. (The computer system controlled dissolver temperature on the basis of the readings from control thermocouples mounted in the heater assembly and not the actual internal temperature of the dissolver vessel; the dissolution temperature of 195°C corresponded to a heater block temperature of 450°C.)

Heatup of the dissolver vessel was continued until the LZ heater temperature exceeded 400°C, the UZ heater temperature (if used) was greater than 375°C, and the dissolver pressure was at least 115 psig. When these conditions were met, the computer logged the "primary dissolution start time" and automatically proceeded to the next procedure step. In this step, the primary dissolution sensors were displayed and updated, keeping the operator apprised of system temperatures and pressures as well as condenser cooling parameters such as cooling water flow rate and temperature differential ("delta T"). After four hours with the dissolver at temperature and pressure, the heaters were de-energized automatically, and sensor readings were displayed to monitor cool down of the vessel. The end time of the primary dissolution, i.e., the heater cutoff time, was also logged.

When the LZ temperature reached 95°C, the process-control state that vented the dissolver to the off-gas hold tank was automatically energized, and the pressure was allowed to equilibrate. Then, the program twice repressurized the dissolver with 120-psig air and vented it to the hold tank to enhance transfer of the dissolver off-gas to the hold tank. Next, the residual gas in the dissolver freeboard was pumped to the hold tank until the dissolver pressure reached 0 to 2 psig. This sequence of steps ensured virtually complete transfer of the off-gas (including the fission-product gases released during dissolution) to the hold tank and thereby assured representative sampling of the gas for the fission-gas determination.

The computer next instructed the operator to put the BT into the "Transfer Weighing Condition" so that solution could be transferred from the dissolver vessel to the BT. In the Transfer Weighing Condition, all BT lines and cables were disconnected except for the (flexible) connections to the solution-transfer piping and to the vacuum reservoir system; this allowed for free movement of the BT during the subsequent dynamic weighing operation.

When the operator acknowledged that this instruction was carried out, the program proceeded to an automated solution-transfer sequence in which the BT was first evacuated and lowered onto the BT balance, the BT tare weight was recorded, and then the dissolver was vented to the cell atmosphere to transfer the dissolver solution into the BT. The computer monitored the pressure in the BT and the vacuum reservoir and continued the solution transfer until this pressure exceeded -0.5 psig, i.e., until vacuum in the reservoir was exhausted. At this point, the computer read the BT balance, calculated the weight of solution transferred into the BT, and compared this weight with a theoretical solution weight based on the weight of the canned segment, the weight of Thorex added, and the estimated weight of cladding hulls that the segment contained (the hulls were presumed not to dissolve, although, in actuality, hull dissolutions to the extent of 10 to 25% occurred). If the weight of transferred solution accounted for at least 98% of the theoretical weight, the transfer was judged complete, and the percent solution transferred was automatically logged. If the transfer did not provide at least 98% of the theoretical weight, the program automatically repeated the transfer sequence after a 15-min delay to allow decay of ^{220}Rn that might have accumulated in the vacuum reservoir tank. Incomplete recovery after the second try resulted in a prompt to the operator to investigate the cause of the low recovery and take necessary remedial action.

After transfer of the dissolver solution to the BT, the program proceeded directly to reflux rinsing of the dissolver vessel, which was done in a manner completely analogous to the contacting with Thorex. The operator was instructed to dispense the appropriate weight (759 g) of nitric acid rinse solution; the identity, batch number, and weight of solution were logged; the reagent was automatically charged to the dissolver through the spray ring; and then a fully automated heatup, refluxing (lasting 15 min), and cool down of the vessel were executed. The rinse solution, once cooled, was also transferred automatically to the BT as described above for the dissolver solution. In the case of the rinse solution, the theoretical solution weight for the material balance calculations included only the weight of rinse solution that had been added to the dissolver.

The next step in the operating procedure depended on the type of segment being dissolved. For fuel-bearing segments, the operating program continued to the sampling of the collected off-gas (Section IV.E.3.c) and then to the secondary dissolution. For plenum segments, the off-gas was not sampled and the second dissolution was omitted, so that the program branched off to the cold rinse of the dissolver vessel, as described below, at the end of the secondary dissolution sequence.

(2) Secondary Dissolution

For the secondary dissolution, one liter (about 1370 g) of Thorex-A1 reagent was added to the dissolver vessel through the dissolver drain line (as opposed to through the internal spray ring) in such a way that the filter in the solution-transfer line was backflushed. This backflushing carried undissolved fuel particles from the filter back to the dissolver vessel.

The secondary dissolution occurred under the same conditions as the primary dissolution but was maintained at pressure and temperature for three hours rather than four. Following the secondary dissolution cool down, just prior to the transfer of the dissolver solution, the computer prompted the operator to prepare for sampling the solution. The operator was instructed to connect a clean, new, disposable sampling line to the sampling port on the solution transfer system and to obtain the two sample bottles designated for secondary dissolution samples. The automated solution transfer was then begun but was automatically interrupted when 500 g of solution (~40% of the total) had been transferred to the BT. At this point, the operator was prompted to obtain samples of the dissolver solution and to weigh the full sample bottles, and the computer recorded the bottle weights. The computer then calculated the net weight of solution in each bottle, recorded this information, and completed the solution transfer. In calculating the percent of the secondary dissolution solution that was recovered in the transfer to the BT, the OPRDSx program included the sum of the net solution weights in each sample as part of the recovered solution. Secondary dissolution samples were not taken for plenum or blank dissolutions.

The above transfer was followed by a 200-mL (hot) reflux rinse with dilute nitric acid and a 250-mL cold rinse; in the latter case, the rinse solution was added to the dissolver vessel through the spray ring and then transferred to the BT without having been heated. At this point, all primary and secondary solutions were present in the BT.

c. Sampling of Dissolver Off-Gas

The off-gas collected during dissolution of each fuel-bearing segment was sampled to determine its fission gas content; other selected components (e.g., NO_x , H_2) were also measured. Off-gas from the dissolution of segments from rod R, the last rod to be processed in the campaign, was not sampled, owing to difficulties with the gas-handling equipment in the DDS hot cell. As a result of frequent failure of the metal-bellows pumps used to transfer and recirculate dissolver off-gas during DDS operations (either through fatigue of the metal bellows or failure of the pump motors), the inventory of spare pumps and spare parts for the pumps was depleted when a failure occurred during processing of rod Q. This failure prompted the installation of a lighter-duty pump, which performed adequately for gas transfers but had trouble recirculating the gas to mix it prior to sampling. Procurement of parts to repair the heavy-duty pump was ruled out because of the long delivery time quoted on the parts and the short time until the end of the campaign. Because of concern that the light-duty pump might fail under the load of gas recirculation and leave the DDS operations without any gas-handling capabilities, ANL asked that BAPL waive its request for sampling and analysis of the off-gas from dissolution of the rod R segments; BAPL acceded to this request.

In general, samples were taken only of the gas from the primary dissolution, in which the bulk (90 to 99%) of the segment fuel was dissolved. Earlier experiences showed that the fission-gas content of the secondary dissolution off-gas represented a negligible fraction of that obtained from the primary and did not merit the effort and cost of its measurement. Before sampling, the gas collected in the off-gas system was thoroughly mixed by recirculating the gas through a closed piping loop that included the off-gas hold tank.

The gas recirculation was automatically initiated by OPRDSx when the operator acknowledged acceptance of the results from the steps just prior to this part of the procedure, i.e., the reflux-rinse solution transfer at the end of the primary dissolution. The gas in the off-gas system was first recirculated through the ~60-L in-cell portion of the loop for 30 min at a 25-L/min flow rate and then was diverted through the sample cylinders attached at the out-of-cell gas-sampling station. Recirculation was continued for a period of 5 min at a flow rate of 20 L/min to flush and fill the cylinders. The two sample cylinders were then isolated, closed, removed from the sampling system, and submitted for analysis. Data pertinent to subsequent calculations of results of the analyses, e.g., the pressure in and temperature of the off-gas hold tank at the time of sampling, were automatically recorded in the process log and the dissolver data summary file.

The gas that remained in the system was slowly vented to the cell air-exhaust system to be diluted with cell air and released to the cell exhaust stack. In carrying out this venting, OPRDSx first displayed the P&I diagram for the off-gas system, energized the appropriate process-control state to vent the off-gas hold tank, logged the time as the start time for off-gas release, and monitored the change in hold-tank pressure as a function of time. The operator was instructed to adjust, as required, the metering valve that controlled the rate of venting to maintain an appropriately low rate of release. When the hold tank pressure reached 1 psig (usually a period of about 2 h), the end time for off-gas

release was automatically logged. Off-gas releases in the DDS were also monitored on the radiation-monitoring system for the exhaust stack.

Except for its not being sampled, dissolver off-gas collected during the secondary dissolution or during dissolution of a plenum segment was treated in identical fashion to that just described.

d. Blend Tank Operations

The next major section of the OPRDSx procedure dealt with weighing, blending, and sampling of the solution in the BT. The solution and rinses accumulated in the BT were blended to provide a homogeneous solution, sampled, spiked with a known amount of NBS 950a standard reference material, and sampled again. The BT assembly and the solution samples were weighed to obtain data required for subsequent analytical calculations and to monitor the material balance during each BT operation.

For each weighing, the BT assembly was first put in the "Standard Weighing Condition" by disconnecting all attached lines, cables, etc., and verifying that it was free of extraneous objects. In this condition, the measured assembly weight could be compared directly to the assembly tare obtained earlier [Section IV.E.3.a.(3)].

The four major BT operations are described below, in the order in which they were carried out.

(1) Blending and Sampling of Unspiked Solution

Upon acceptance by the operator of the computer-calculated material balance, after transfer of the final (cold) rinse that followed the secondary dissolution, OPRDSx proceeded directly into the BT operations, which started with an initial weighing of the unspiked, unmixed, unsampled (UUU) solution. The assembly was then automatically weighed in triplicate to the nearest gram. The automated sequence of operations included raising and lowering the assembly off of and onto the balance for each weighing by means of air-powered pistons on the BT support structure, reading the balance, averaging the results, calculating a net solution weight by subtracting the BT assembly tare weight, and displaying the individual weight values for operator acceptance. When accepted by the operator, all the weighing data were logged automatically.

Next, the solution in the BT was mixed by stirring it for 30 min with the integral stirrer built into the BT assembly. The OPRDSx program prompted the operator through the steps involved in connecting appropriate hoses to the BT, coupling the stirrer to the stirrer motor, and initiating the mixing operation. During the mixing step, the program monitored and displayed the elapsed mixing time, remaining mixing time, vacuum-reservoir pressure and pressure-increase rate, and the pressure in the BT. Any off-normal sensor readings caused an alarm message to be displayed, which instructed the operator to take remedial action. During the mixing step, a slow flow of cell air was drawn through the BT headspace into the vacuum reservoir tank to allow for decay of ^{220}Rn that emanated from the solution. This precaution prevented this gaseous, alpha-emitting radionuclide from being released to the cell atmosphere.

At the end of the mixing period, the BT assembly was automatically weighed to provide a weight of the unspiked, mixed, unsampled (UMU) BT solution. The OPRDSx program calculated this solution weight from the UMU assembly weight and the assembly tare and also calculated the weight of solution lost (as vapor) from the BT during mixing by subtracting the UMU solution weight from the UUU solution weight recorded earlier. Generally, the weight loss on mixing was smaller than could be determined with the BT balance (i.e., less than 1 g). All the measured data and calculated weights were displayed for operator acceptance and logged. The UMU weight of solution in the BT was taken, in subsequent analytical calculations, to be the most representative weight of solution in the unspiked BT.

In the next step, samples of the UMU BT solution were withdrawn for measuring the isotopic composition of uranium in the segment and assaying the segment for fission-product monitors (^{137}Cs , ^{144}Ce , and ^{95}Zr). Four separate samples of the solution were taken in the case of fuel-bearing or plenum segments; only two were taken for blank runs.

In sampling the BT solution, the operator was first prompted to obtain the preweighed sample bottles designated for the unspiked BT (UB) samples and to install a new, clean, disposable sampling assembly in the BT solution-sampling system. In addition, a new, clean, disposable, polyethylene sleeve was placed in the orifice of the gate-type access valve on the BT to prevent contamination of the valve's internal surfaces by droplets of solution that might adhere to the in-tank portion of the sampling line when the line was later removed from the tank. The operator was then prompted to start the BT mixer (the solution was stirred continuously during sampling) and obtain the appropriate number of samples, each about 25 mL.

After the samples were taken, OPRDSx prompted the operator to remove and discard (to waste) the sampling assembly and polyethylene sleeve and to cap and weigh the individual sample bottles. The bottle weighings were made in triplicate to the nearest 0.001 g. From the weighing data, the computer calculated a net solution weight for each sample, using the previously recorded tare for each sample bottle. Next, the BT assembly was automatically weighed as before. The net weight of solution in the BT was calculated and recorded as the weight of the unspiked, mixed, sampled (UMS) BT solution. The computer also calculated the weight of solution lost during sampling by comparing the UMU solution weight with the sum of the UMS solution weight and the net weights of solution in the several sample bottles. This lost solution, usually attributable to the small droplets of liquid that adhered to the inside of the tubing in the discarded sampling assembly, generally amounted to 2 g or less. All the information from these weighings and calculations was displayed for operator acceptance and, once accepted, logged. The program then proceeded to the next BT operation, spiking of the BT solution.

(2) Spiking of the Blend Tank Solution

The BT solution that contained each fuel-bearing segment was spiked in a manner that ensured verifiable transfer of the spike into the BT, complete spike dissolution, and thorough homogenization and isotopic equilibration of spike and segment uranium in the solution. The BT spiking operations were

automatically omitted from the DDS procedure for plenum dissolutions and blank runs because the uranium content of the BT solution was small in these cases and did not need to be determined with high precision and accuracy.

The spike added to the BT solution was an accurately weighed quantity of NBS Standard Sample 950a (a natural uranium assay standard in the form of U_3O_8), contained in a sealed nickel capsule. For QA purposes, the preparation, storage, and dispensing of the spikes followed strict guidelines. The spikes were prepared in batches by the ACL according to the procedure in Document C-0030-289 (Attachment 1) and were stored in labeled, closed glass bottles in a locked cabinet in one of the ACL laboratories. At the onset of DDS operations with segments from a given fuel rod, the group of spikes designated for use with that rod was transferred to the DDS task leader and stored in a safe in the DDS work area.

Each fuel-bearing segment to be dissolved in the DDS was assigned a particular spike by means of an entry in the FRIF. Two sizes of NBS 950a spike were used for the EOL campaign with the aim of keeping the spike/sample ratio for most segments within the optimum 1:5 to 5:1 range for subsequent mass spectrometric measurements on uranium in the spiked BT solution: one size contained a nominal weight of U_3O_8 equivalent to 1 g of uranium and the other, a nominal weight equivalent to 3 g of uranium. The 1-g spikes were used for segments having a uranium loading of less than 2.5 g, and the 3-g spikes were used for segments containing more than 2.5 g of uranium.

In executing the BT spiking operations, the OPRDSx program first retrieved, from the FRIF, the spike identification assigned to the segment being processed and directed the operator to obtain the spike from the shift supervisor and transfer it into the cell enclosure. Next, the operator was prompted to weigh the spike, including the bottle that held it, to verify the spike identity. The computer compared this spike weight with a corresponding weight recorded in a computer file at the time the spike was prepared by the ACL. If the weights agreed within preset limits, the program proceeded to the next step. Otherwise, the operator was required to resolve the discrepancy and repeat the weighing before the program would continue.

Once the spike identity was verified, the operator was prompted to charge the spike to the BT by gently "pouring" the nickel capsule into the (funnel-shaped) opening of the BT access valve. This method of addition of the spike avoided touching the spike capsule with possibly contaminated M/S manipulator fingers and thereby contaminating the BT solution with extraneous uranium. The operator verified that the spike had passed through the access valve and into the tank by visual inspection of the orifice, using a mirror positioned over the valve, and also verified that the BT balance had registered a weight increase upon addition of the spike.

Next, the BT mixer was started and a slow flow of cell air through the BT headspace was established by a procedure analogous to that described for mixing of the unspiked BT solution. The program next proceeded to a BT heatup sequence wherein the BT solution temperature, vacuum-reservoir pressure, BT pressure, and BT external temperature were read and displayed every 30 s. The computer also monitored the solution heatup rate and the rate of pressure increase in the vacuum reservoir and prompted the operator to make appropriate adjustments if these rates were outside preset tolerances.

The heatup sequence continued until the solution temperature reached 95°C, at which time the program initiated the "spike dissolution" routine. The start time of the spike dissolution was recorded, and the solution temperature was held between 95 and 105°C for 60 min. Monitoring of appropriate temperatures, pressures, and rates of change continued through the dissolution routine, and messages to the operator calling for appropriate adjustments of the heating rate or air flow were displayed as required. When 60 min at temperature had elapsed, the spike-dissolution end time was automatically logged, and the operator was prompted to turn off the BT heater and mixer and to place the BT assembly in the standard weighing condition.

The BT assembly was then weighed automatically as previously described; the net solution weight obtained from these weighings was recorded as the spiked, mixed, unsampled (SMU) BT solution weight. The SMU solution weight was then used, with the UMS weight determined previously, to calculate a value for any loss of weight during the spike dissolution. All of the weighing data was displayed for operator acceptance and logged. Weight loss during spiking of the BT generally amounted to a few (less than ten) grams and was attributable to evaporation of the hot solution. When the SMU weight data were accepted by the operator, the program proceeded to sampling of the spiked BT solution.

(3) Sampling of the Spiked Blend Tank Solution

To make efficient use of operator time during the BT operations, the spiked BT solution was not cooled prior to being sampled. Cooling the solution at this point in the procedure would have required connecting cables and fittings to the BT and then disconnecting them to regain the standard weighing condition. When performed with M/S manipulators, such operations are time-consuming. Sampling the solution while it was still warm entailed no significant disadvantages, although the solution weights obtained may not have been as reliable as if the solution were cooled first, because of larger vapor losses taking place. The weights of the spiked BT solution and solution samples are of operational interest only and are irrelevant to subsequent analytical calculations (see Section V.E); thus, they did not need to be determined as accurately as, for example, the corresponding data on the unspiked BT solution.

Four separate samples of the spiked BT solution were weighed by a procedure analogous to that followed during sampling of the unspiked BT solution. The net weight of solution in the BT after removal of the samples--designated the spiked, mixed, sampled (SMS) BT solution weight--was added by the computer to the sum of the net solution weights for the spiked BT solution samples and was automatically compared with the SMU solution weight to give a value for the solution lost during sampling the spiked BT. In general, this calculated weight loss (in the range 2 to 5 g) was somewhat larger than that found for sampling the unspiked BT, probably as a result of evaporation of the warm solution. All of the weight data obtained during the spiked BT sampling were displayed for operator acceptance and logged in appropriate computer and hardcopy files.

(4) Transfer of Blend Tank Solution to Waste Storage Tank

When the foregoing operations had been completed, OPRDSx prompted the operator to initiate cooling of the BT solution and then proceeded

to direct the operator through a series of post-dissolution operations (described in the next subsection) while the solution cooled. When the BT solution temperature reached 60°C or less, the operator was prompted to tilt the BT by means of the stanchion support mechanism and to implement transfer of the solution to the waste-storage tank designated in the FRIF for the given segment. This transfer was made by pumping the solution out of the BT through a portable plastic drain line inserted through the BT access valve. Tilting the BT while it was being drained resulted in the removal of all but a few grams of solution from the tank. Following computer prompts, the operator then rinsed the BT with 200 g of dilute nitric acid and transferred the rinse solution to the same waste tank. After the rinse solution was removed, the BT was automatically weighed, and the weight of solution remaining in the BT was calculated as the difference between this weight and the empty BT weight recorded at the beginning of the dissolution. If the remaining solution weight exceeded 20 g, the program automatically repeated the prompts that directed draining of the tank. This requirement was included in the procedure because large amounts of fuel-bearing solution left in the BT at this point hampered the effectiveness of subsequent BT decontamination procedures. During these operations, the computer monitored and recorded readings from the waste-tank level probes, as well as documented the weighing data and other pertinent information such as reagent batch number and weight of the rinse reagent. The empty, but still contaminated, BT was set aside for later decontamination [Section IV.E.3.f.(1)] at a convenient time in the DDS operations schedule.

e. Post-Dissolution Operations

Following completion of the BT operations in each dissolver run, the operator was prompted through a series of operations to restore the equipment and work areas to a condition appropriate for beginning the next run. These operations are listed below, with a brief description of the purpose of each.

(1) Flushing the Off-Gas Hold Tank

The off-gas hold tank was flushed with air (30 min at a flow rate of 5 to 10 L/min) to displace off-gas components from the run just completed.

(2) Inventory of Undissolved Cladding Hulls

To cross-check the identity of the segment charged to the dissolver and to obtain information regarding the extent of dissolution of the segment cladding, we made an inventory of the undissolved cladding hulls, as follows. The dissolver vessel was opened, the fuel basket was removed, and the cladding hulls remaining in the basket were individually inspected and counted and then weighed as a group. The computer compared the hull count with the number of hulls that would be expected, based on the number of cuts made in shearing the segment. A recount was done if the numbers disagreed. If the discrepancy persisted, the operator was required to enter any observations made during the counting. The DDS operators counted only hulls that were recovered intact and did not attempt to piece together hulls that might have been recovered as fragments. In general, discrepancies between the number of hulls expected and recovered were explained by the presence of cladding fragments among the hulls.

The OPRDSx program calculated an estimate of the weight of cladding that had dissolved, using the measured hull weight and the cladding-weight estimate, as found in the FRIF, for the appropriate segment.

(3) Removal of the Filter-Housing Assembly

The filter-housing assembly was removed from the solution-transfer system, and its end-fittings were capped to avoid spreading residual solution while the housing was being transferred to the in-cell maintenance area. The assembly was subsequently decontaminated and disposable components were replaced to prepare it for reuse in the dissolver system.

(4) Treatment of Discarded Sampling Assemblies

Discarded sampling assemblies were rinsed with distilled water to reduce residual levels of radioactivity and were cut into small pieces to reduce the volume they occupied in the waste containers that would be transferred out of the cell. This treatment reduced radiation levels at the surface of the containers and increased the payload, which in turn reduced disposal cost.

The rinse solution was concentrated by evaporation in a heated vessel; periodically, the concentrate was added to the liquid-waste storage tanks for disposal with other liquid residues from the DDS operations.

(5) Check for Liquid in the Off-Gas Hold Tank

A sampling bottle was placed under the drain line for the off-gas hold tank, and the drain was opened to check for liquid that might have accumulated in the off-gas system. Such liquid could arise from solution droplets entrained with the dissolver off-gas or from vapor that condensed in the tank. This check was performed routinely in the early portion of the EOL campaign, but was discontinued after processing of segments from several rods showed that no liquid accumulated in the hold tank under normal conditions. It was reinstated as a routine operation after a mishap with a later segment (J-03) showed that, under certain conditions, dissolver solution could be carried into the off-gas system without the operator being aware of it. If liquid was found in the off-gas hold tank, it was submitted for analysis, and appropriate actions were taken to recover any fuel material associated with it to account for this fuel in the segment assays.

(6) General Housekeeping

General housekeeping was performed to ensure that all work-area tools were returned to their designated storage locations, and that the work area was clean and free of unnecessary clutter. As part of this effort, the stainless-steel drip pan under the dissolver control-valve bank was rinsed with dilute nitric acid and wiped down to minimize accumulation of fuel material on the tray surfaces. Because of this cleaning after each dissolver run, solution that collected in the tray from leaking valves or transfer lines could be recovered without fear of gross contamination of the recovered solution by extraneous fuel material from previous runs. Such leakage occurred on several occasions during the EOL campaign but was successfully dealt with in every case (see Section V.C.3.b), partially due to the good housekeeping practiced in the cell work areas.

f. Decontamination of Reusable Dissolver Components

Wherever practical, small equipment items used in analytically sensitive operations (e.g., sample bottles, sampling lines) were used once and discarded to prevent cross-contamination of fuel material or samples from one segment with those from another. However, economic considerations required that individual components be decontaminated and reused for three kinds of items: the BTs, the filter housings for the solution transfer system, and the gas-sampling cylinders. This section outlines the decontamination procedures employed for each type of item.

(1) Blend-Tank Decontamination

Special care was taken to remove residual fuel solution from each BT between uses and to ensure that the tank was thoroughly dry before reuse. These precautions were necessary to prevent contamination of the BT solution from one segment with fuel from another and to ensure a valid net weight for the solution collected in the BT during a given segment dissolution. (If liquid is present in the "empty" BT when its tare weight is determined, the net weight of solution in the tank, as determined by subtracting the tare from a subsequent assembly weight, will be lower than the actual solution weight by the amount of this extraneous liquid.)

To achieve these conditions, each BT was taken through a decontamination procedure that included two separate refluxes of dilute nitric acid in the tank and a drying step wherein the tank was heated while being flushed with a stream of air to displace vapor in the tank. The decontamination procedure was carried out under control of a subsection of the OPRDSx program. This subsection could be initiated as part of the post-dissolution operations that immediately followed dissolution of a given segment or could be run separately at a later time. If run separately, the procedure was treated as a continuation of the OPRDSx run for the segment dissolution for which the BT was last used. Thus, records related to the BT decontamination were kept together with records of other data from that segment dissolution.

For each reflux rinse of the BT, 250 mL of dilute nitric acid was charged to the tank, heated to 95°C, cooled to 70°C, and removed by means of the BT sampling system (with clean, new sampling assemblies sized to fit appropriate sample bottles). The first rinse solution was discarded to the waste-evaporation vessel, while the second one was collected in a new, clean sample bottle and submitted for analysis. The BT was dried by heating to an internal temperature of 120°C under a controlled air flow for 30 min. The dried BT was cooled, weighed, and placed in storage pending the results of analysis of the second rinse solution. The dry BT weight was automatically compared with the empty BT weight recorded at the beginning of the dissolver operations. The weight after decontamination had to be less than the recorded empty weight plus 2 g or the OPRDSx program directed the operator to repeat the drying step or take other remedial action as appropriate. If the dry BT weight was acceptable, the weight was logged in the segment dissolution data records and in the HISLOG file for the BT.

The second reflux rinse solution was analyzed for total uranium as described in Section IV.F.1. For decontamination of the BT to be judged satisfactory, uranium in the rinse solution had to fall below 100 µg, a

value that pre-EOL testing had shown to correspond to a residual level of 10 μg or less uranium in the BT. This quantity of residual uranium, in turn, corresponded to an amount of fuel in the BT that would have a negligible impact on the composition of fuels from other segments subsequently collected in the BT. The results of analysis of the individual BT rinses were reported by the ACL to the DDS supervisor, who entered them into the HISLOG files for each BT. If the results indicated satisfactory decontamination, the supervisor also entered a code indicating approval for reuse of the tank.

(2) Decontamination of Filter-Housing Assemblies

In spite of the difficulty associated with handling small parts with M/S manipulators, filter-housing assemblies that became contaminated during dissolver operations had to be decontaminated remotely because of a buildup of ^{125}Sb , ^{95}Nb , and other easily hydrolyzable, gamma-emitting nuclides on the internal metal surfaces of the housings. The mechanism for this buildup has not been identified, but testing early in the EOL campaign indicated that the radioactivity on the metal surfaces could not be removed without drastic chemical cleaning, sufficient to etch the surfaces.

Each contaminated housing was disassembled at the general-purpose work area of the DDS cell. Special jigs and wrenches fabricated for this purpose were used. The prefilter and membrane filter from each unit was removed and placed in a labeled, covered plastic Petri dish, and the radioactivity of the filter media was measured with the portable in-cell radiation probe. Individual filters generally read between 20 and 100 R/h at 4 in. On the basis of gamma-counting data obtained for some of the more-active specimens, activity on the filters was due primarily to ^{125}Sb , with lesser contributions from ^{95}Nb , and only minor contributions from the fission products that were of interest in our work (i.e., ^{137}Cs and ^{144}Ce). The individual sets of filter media were stored in a shielded container within the cell and were disposed of at the end of the campaign by incorporating them into the cement that contained the liquid dissolver residues.

The metal housing of each filter assembly was visually inspected for the presence of undissolved fuel particles (none was ever found during the campaign) and thoroughly rinsed with dilute nitric acid. The rinse solutions were collected and their volume reduced in the waste-evaporation vessel for periodic addition to the waste-storage tanks.

Next, clean, new filter media and O-rings were installed in each housing, and the unit was then reassembled. Each assembly was then pressure-tested by attaching it to the pressure manifold installed at the work station for this purpose and pressurizing the manifold to 120 psig. To pass the test, the assembly had to lose no more than 1 psig over a 1-min period. When an assembly was judged satisfactory, it was removed from the manifold, its ends were capped, and it was placed in a storage area designated for decontaminated filter-housing assemblies. The operator entered into the HISLOG file for each assembly the information pertinent to the decontamination, including the radiation-monitor reading obtained for the used filter media.

(3) Purging of Gas-Sampling Cylinders

After the gas in each sampling cylinder had been analyzed, the cylinders were returned from the ACL to the DDS and connected to the off-gas sampling station. Each cylinder was then purged of residual sample gas by flushing with compressed air for 5 min. After the purging operation, the operator entered the date of purging and his or her identity into the HISLOG files for the appropriate cylinders. The cylinders were then ready for reuse.

4. Data Summary and Management Review of Segment Dissolution

After DDS operations for any given segment, the data were processed with a stand-alone report-generation program, which produced a dissolution data summary as illustrated in Fig. 17. This program read the FRIF for the rod from which the segment was sheared and the summary data file generated during DDS operations with the segment, calculated results from the various measured

Page 1 of 6 DISSOLUTION DATA SUMMARY SEGMENT DISSOLVED: B-01 DATE/TIME RUN STARTED: 21 AUG-84 08:45 AB B-01 DATE/TIME RUN ENDED: 24-AUG-84 08:00 SB B-01 SEGMENT DESCRIPTION ROD DIA: 0.527 NOMINAL CUT LENGTH: 0.250 ESTIMATED CLAD WT: 47.800 ESTIMATED OXIDE WT: 300.878 ESTIMATED TOTAL WT: 848.678 ESTIMATED U CONTENT: 0.125 SEGMENT WEIGHING FULL CAN WT, CELL M-3: 856.918 FULL CAN WT, CELL M-1: 856.920 WT DIFFERENCE: 0.007 EQUIPMENT AND MATERIALS USE DISSOLVER SYSTEM: DS1 BLEND TANK: TK-01 SOLUTION FILTER: FR-01 GAS SAMP CYL 1 10 NO: SC-01 GAS SAMP CYL 2 10 NO: SC-11 MSD-MSDA SPIKE: 60A-121	Page 2 of 6 PRIMARY DISSOLUTION SEQUENCE - SEGMENT B-01 DISSOLUTION REAGENT USAGE THOREX WT SPECIFIED: 411.8 PRI DIS REAGENT USED: THOREX PRI DIS REAGENT WT: 411.8 PRI DIS REAGENT BATCH NO: 04 TIME OF DISSOLUTION PRI DIS START TIME: 19:58:59 PRI DIS END TIME: 20:59:56 SOLUTION TRANSFER TO BLEND TANK PRI DIS SOL XFER WT: 4371 PRI DIS % SOL XFERRED: 98.85 REFLUX RINSE REAGENT USAGE PRI RFLX RINSE REAG USED: NITRIC PRI RFLX RINSE WT: 759 PRI RFLX RINSE BATCH NO: 2 TIME OF REFLUX PRI RFLX START TIME: 08:38:11 PRI RFLX END TIME: 08:54:07 RINSE TRANSFER TO BLEND TANK PRI RFLX RINSE XFER WT: 795 PRI RFLX % RINSE XFERRED: 104.75	Page 3 of 6 SECONDARY DISSOLUTION SEQUENCE, SEGMENT B-01 DISSOLUTION REAGENT USAGE SEC DIS REAGENT USED: THOREX-AL SEC DIS REAGENT WT: 1377 SEC DIS REAGENT BATCH NO: 02 TIME OF DISSOLUTION SEC DIS START TIME: 14:10:48 SEC DIS END TIME: 17:12:06 SOLUTION TRANSFER TO BLEND TANK SEC DIS SOL XFER WT: 1268 SEC DIS % SOL XFERRED: 96.72 REFLUX RINSE REAGENT USAGE SEC RFLX RINSE REAG USED: NITRIC SEC RFLX RINSE WT: 202 SEC RFLX RINSE BATCH NO: 02 TIME OF REFLUX SEC RFLX START TIME: 01:04:48 SEC RFLX END TIME: 01:05:26 RINSE TRANSFER TO BLEND TANK SEC RFLX RINSE XFER WT: 222 SEC RFLX % RINSE XFERRED: 109.91	Page 4 of 6 COLD RINSE REAGENT USAGE SEC COLD RINSE REAG USED: NITRIC SEC COLD RINSE WT: 253 SEC COLD RINSE BATCH NO: 2 RINSE TRANSFER TO BLEND TANK SEC COLD RINSE XFER WT: 256.02 SEC COLD % RINSE XFERRED: 101.19	Page 5 of 6 SAMPLE BOTTLE WEIGHT DATA SEGMENT NO: B-01 <table border="1"> <thead> <tr> <th>BOTTLE NO</th> <th>EMPTY BOTTLE WT IN CELL</th> <th>EMPTY BOTTLE WT DIFFERENCE</th> <th>FULL BOTTLE WT</th> <th>NET SAMPLE WT</th> </tr> </thead> <tbody> <tr> <td>DS-1</td> <td>75.989</td> <td>0.004</td> <td>111.228</td> <td>28.239</td> </tr> <tr> <td>DS-2</td> <td>75.484</td> <td>0.008</td> <td>111.109</td> <td>35.625</td> </tr> <tr> <td colspan="4"></td> <td>SUM DS NET SAMPLE WTS: 63.865</td> </tr> <tr> <td>UB-1</td> <td>77.097</td> <td>0.004</td> <td>111.578</td> <td>34.481</td> </tr> <tr> <td>UB-2</td> <td>76.849</td> <td>0.003</td> <td>111.638</td> <td>34.789</td> </tr> <tr> <td>UB-3</td> <td>75.544</td> <td>0.008</td> <td>114.704</td> <td>39.160</td> </tr> <tr> <td>UB-4</td> <td>75.667</td> <td>0.002</td> <td>110.487</td> <td>34.770</td> </tr> <tr> <td colspan="4"></td> <td>SUM UB NET SAMPLE WTS: 148.201</td> </tr> <tr> <td>SB-1</td> <td>76.110</td> <td>0.002</td> <td>109.889</td> <td>33.779</td> </tr> <tr> <td>SB-2</td> <td>75.571</td> <td>0.003</td> <td>108.775</td> <td>33.204</td> </tr> <tr> <td>SB-3</td> <td>75.656</td> <td>0.004</td> <td>110.482</td> <td>34.826</td> </tr> <tr> <td>SB-4</td> <td>75.746</td> <td>0.002</td> <td>112.487</td> <td>36.741</td> </tr> <tr> <td colspan="4"></td> <td>SUM SB NET SAMPLE WTS: 138.550</td> </tr> </tbody> </table> NOTE: ALL WEIGHTS IN GRAMS.	BOTTLE NO	EMPTY BOTTLE WT IN CELL	EMPTY BOTTLE WT DIFFERENCE	FULL BOTTLE WT	NET SAMPLE WT	DS-1	75.989	0.004	111.228	28.239	DS-2	75.484	0.008	111.109	35.625					SUM DS NET SAMPLE WTS: 63.865	UB-1	77.097	0.004	111.578	34.481	UB-2	76.849	0.003	111.638	34.789	UB-3	75.544	0.008	114.704	39.160	UB-4	75.667	0.002	110.487	34.770					SUM UB NET SAMPLE WTS: 148.201	SB-1	76.110	0.002	109.889	33.779	SB-2	75.571	0.003	108.775	33.204	SB-3	75.656	0.004	110.482	34.826	SB-4	75.746	0.002	112.487	36.741					SUM SB NET SAMPLE WTS: 138.550
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Page 2 of 6 BLEND TANK WEIGHT DATA SEGMENT S-01 BLEND TANK: TK-01 OPERATION INITIAL WEIGHING ASSEMBLY WT: 34401 SUPPORT WT: 27074 EMPTY BLEND TANK WT: 7327 UNSPIKED BLEND TANK WEIGHING WT, BEFORE MIXING, BEFORE SAMPLING: 41301 WT, AFTER MIXING, BEFORE SAMPLING: 41300 SOLUTION CHNG FROM MIXING OPERATION: -1 WT, AFTER MIXING AND SAMPLING: 41167 SOLUTION REMOVED DURING SAMPLING: 8765 SOLUTION LOSS DURING SAMPLING: 148 SOLUTION LOSS DURING SAMPLING: 0	Page 3 of 6 OFF-GAS DATA SEGMENT: B-01 GAS SAMPLING CYLINDERS USED GAS SAMP CYL 1 10 NO: SC-01 GAS SAMP CYL 2 10 NO: SC-11 GAS SAMPLING PARAMETERS HOLD TANK PRESSURE: 17.5 HOLD TANK TEMP: 23.8 GAS RELEASE PRI OFF-GAS RISE STR TIM: 10:38:54 PRI OFF-GAS RISE END TIM: 10:55:04 SEC OFF-GAS RISE STR TIM: 18:54:02 SEC OFF-GAS RISE END TIM: 14:04:11	Page 4 of 6 HAUL DATA SEGMENT: B-01 ESTIMATED CLAD WT: 47.820 HAUL WT: 86.831 DISSOLVED CLAD WT: 11.488 CLAD % DISSOLVED: 24.0 NO OF HAULS FROM SHEAR: 44 NO OF HAULS RECOVERED: 44	Page 5 of 6 SPIKED BLEND TANK WEIGHING WT AFTER SPIKING, BEFORE SAMPLING: 41180 SOLUTION CHNG FROM SPIKING OPERATION: -7 WT AFTER SPIKING AND SAMPLING: 41018 SOLUTION REMOVED DURING SAMPLING: 6611 SOLUTION LOSS DURING SAMPLING: 138 SOLUTION LOSS DURING SAMPLING: 0 BLEND TANK ASSEMBLY WT AFTER DRAINING: 34400	Page 6 of 6																																																																						

Fig. 17. Example of Dissolution Data Summary Report

parameters, and wrote the results to a new file in a format appropriate for management review. As Fig. 17 shows, the summary has seven major parts: (1) data that characterize the segment and codes that identify equipment and materials used in dissolution, (2) data pertinent to the primary dissolution sequence, (3) a similar listing for the secondary dissolution, (4) data related to the off-gas sampling operations, (5) data regarding the recovered cladding hulls and extent of cladding dissolution, (6) sample bottle weights, and (7) weights obtained during BT operations. The items included in each section of the summary are self-explanatory and will therefore not be discussed individually.

The dissolution data summary for each segment was reviewed by the DDS task leader, the project principal analyst, and the project manager to ensure completeness of the data listings and to identify any operational or processing deviations that might have been indicated by the data. Omissions or inconsistencies were dealt with on a case-by-case basis. In general, missing data were retrieved by the DDS task leader from the hardcopy process log or from the manual logbook maintained at each dissolver work station, and were added to the dissolver data summary file by means of a special data-entry program. This program not only wrote the new information to the summary file, but also noted in the file that it had been edited, and indicated the identity of the editor and the date. If the new data replaced previous entries that were judged to be in error during the post-dissolution review, both the old and new information were retained in the summary data file, although the program that formatted the data to produce the dissolution data summary used only the most-recent entry in producing the report. This system for reviewing data from the segment dissolutions provided needed flexibility in the handling of the computerized data records and, at the same time, maintained strict traceability of the recorded information for QA purposes.

5.1 Archive Storage of Solution Samples

To satisfy the project requirement for taking and retaining samples of fuel solution for each segment, half of the samples of each solution type [i.e., secondary dissolution (DS), unspiked BT (UB), and spiked BT (SB)] were placed in archive storage, while the other half were transferred from the DDS to the Analytical Hot Cell for chemical and radiometric analysis. Specific samples of each type were designated for each purpose. From the two DS samples obtained for fuel-bearing segments, the first (the DS-1) sample was archived, and the second (the DS-2) was submitted for analysis. We reasoned that the DS-2 sample was less prone to contamination, since taking of the DS-1 sample rinsed the solution transfer-system sampling port, and would provide a more-representative primary analysis of the secondary dissolution. Among the four samples obtained from each type of BT solution, the first and last (i.e., UB-1 and UB-4 or SB-1 and SB-4) were analyzed and the middle two (i.e., UB-2 and UB-3 or SB-2 and SB-3) were archived. Duplicate samples of each BT solution were analyzed to assess sampling errors arising from inhomogeneity of the BT solution. Because the BT solution was continuously mixed during sampling, we expected that such inhomogeneity would have its greatest impact on a comparison between the first and last samples taken. These were, therefore, the samples we selected for immediate analysis. Moreover, the pair of archive samples and pair of analytical samples selected in this way were both symmetrically taken about the midpoint of the sampling operations and shared a common average sampling time with the entire group of four samples. As a consequence, the average of

each analytical result obtained for one pair was expected to be comparable to that of the other pair and to that of the whole group.

To aid the DDS operators in selecting individual samples for transfer to storage or to the Analytical Hot Cell, different-colored labels were used for the bottles. Labels on the archives samples were yellow, the other white.

The archives samples were placed in special bottle holders inside sealable SS cans of the type used with the can-out intercell transfer system. Each can held 12 bottles, and when full, it was transferred to one of the shielded storage tubes located in the floor of the corridor adjacent to the DDS cell. Detailed records were kept with regard to the identity of the bottles in each can and the location of each can in the storage tubes so that individual bottles could be readily retrieved if required. Approximately 800 bottles containing archives solution samples were accumulated during the EOL campaign.

F. Analytical Operations

Operations performed by the ACL on behalf of the LWBR-POB project included (1) preparation of reagents and standards for the DDS operations and (2) analysis of solution and gas samples from operations in the FSSF and DDS. Each step in these analytical operations was performed in accord with a written or computerized procedure that guided a qualified analyst through appropriate preparative chemistry or measurement methodology. A listing of the written procedures employed in these operations is presented in Attachment 1.

Preparation of reagents (Thorex, Thorex-A1, and dilute nitric acid rinse) for use in the DDS was described in Section IV.E.3.b., and the NBS 950a spikes prepared for addition to the dissolver blend-tank solutions were described in Section IV.E.3.d.2. In the following sections, we summarize the procedures that were employed in analyzing the dissolver-solution samples and briefly describe the analysis of gas samples by mass spectrometry.

1. Chemical and Radiometric Analysis of Dissolver Solutions

a. Overview

Selected solution samples obtained during dissolution of each segment were transferred from the dissolver cell to the Analytical Hot Cell by means of the radio-controlled MULE. When the set of samples from a given segment was transferred, a computer file, designated the DDS information file or DDSIF, was automatically generated in the project computer system. This file summarized data pertinent to each solution sample as recorded during the segment dissolution, including (1) the full and empty weights of each sample bottle (samples submitted for analysis as well as those placed in archive storage were listed), (2) the weight of solution contained in the unspiked blend tank (UBT) for the specified segment, and (3) estimates of the total oxide weight and total uranium loading of the dissolved segment as provided by BAPL in the shearing plan for the corresponding fuel rod. The data in the DDSIF were used by computer programs that controlled subsequent aliquoting and analysis of the samples to perform such tasks as verifying the identity of the samples, applying

corrections for evaporation of the solutions, and specifying optimum aliquot sizes for the analysts to take for each designated analysis.

For every fuel-bearing segment, the samples received for analysis included one sample of the secondary dissolver (DS) solution, two separate samples of the UBT solution, and two separate samples of the spiked blend tank (SBT) solution (cf. Section IV.E.5). A weighed aliquot of the DS sample was spiked with NBS 993 (primarily ^{235}U) and was analyzed to determine its $^{233}\text{U}/^{235}\text{U}$ ratio for use in computing, by isotope dilution formulas, the concentration of segment uranium in the DS solution. Each of the two UBT samples was divided by weight into four aliquots to separately measure (1) the segment uranium isotopy, (2) the uranium concentration in the UBT solution, (3) the concentrations of the fission products ^{137}Cs and ^{144}Ce , and (4) the concentration of the fission product ^{95}Zr . One aliquot from each of the SBT samples was prepared to determine the $^{233}\text{U}/^{238}\text{U}$ isotope ratio in the SBT solution and thereby allow calculation of the amount of segment uranium present in the SBT. Our approach of taking a single aliquot from each of two separate samples of the BT solutions provided replicate data with which we could evaluate the reproducibility of the entire sampling, sample preparation, and measurement sequence. These data are used in Appendices B, D, and E to assess the quality of such operations as mixing of the BT solution, as well as to obtain a measure of random errors associated with the results of the individual assays that were performed on the dissolved segments.

In contrast to the fuel-bearing segments, only two dissolver solution samples were analyzed from dissolution of each fuel rod plenum section. These were duplicate samples of the UBT solution (the BT was not spiked during plenum dissolutions), which were assayed for the ^{233}U and ^{232}Th concentration by mass spectrometric isotope dilution (MSID). In general, uranium in the dissolved plenums was at such low levels as to preclude meaningful measurement of its isotopy. On a few occasions, however, uranium in a specific rod plenum was at a level (concentration in the UBT solution greater than $10\text{ }\mu\text{g/g}$) that allowed its isotopy to be determined. In these cases, an additional aliquot from each UBT sample was taken (subsequent to obtaining the assay results) and prepared for isotopic analysis.

During dissolution of a few segments, there were mishaps (see Section V.C.3.b) which resulted in additional samples being generated as part of actions taken to recover. When these special samples took the form of solutions, they were treated in a manner similar to the UBT solutions. Often, however, the samples were submitted as tissues used in wiping down specific areas of the dissolver equipment or hot cell. These tissues were thoroughly leached with nitric acid, and the leach solutions were treated in the same way as were the other special samples.

Each solution aliquot designated for a particular type of measurement or analysis was prepared in a manner to provide a suitable sample. In cases where uranium isotope ratios were to be measured (the case for both uranium isotopic measurements and assays), uranium in the aliquot was isolated and purified for subsequent mass spectrometric or alpha pulse height analysis (APHA). Aliquots designated for fission-product assays by gamma spectrometry were diluted to a uniform volume consistent with the geometry requirements of the counting techniques that were employed.

The individual analytical operations that were executed in aliquoting the dissolver solution samples, preparing the aliquots for analysis, and performing measurements on the prepared aliquots are summarized in the following sections. Results of these measurements generally had to be combined with each other or with data from other project operations to derive information pertinent to the dissolved segment. Calculation of the segment-related parameters and estimation of associated errors are discussed elsewhere in this report, as follows:

Uranium Isotope Abundances	Section V.D and Appendix B
Uranium Assay Results	Section V.E and Appendix D
Fission-Product Assay Results	Section V.F and Appendix E

b. Aliquoting of Dissolver Solution Samples

The taking of weighed aliquots from the dissolver solution samples represented a critical first step in the methodology by which measurements on the samples were performed. As such, a substantial level of quality control was incorporated into the procedures that were established for these operations. Insofar as was practical, operations, data taking, calculations, and data storage were carried out under computer control through a stand-alone program, K3ALQ. This program provided "prompt-and-go" instructions to guide the analyst through operating sequences that required consistent or predetermined ordering of steps to be executed. It anticipated sample numbers and spike identifiers by referring to computer files containing pertinent information, and created files of information and data regarding each sample or aliquot that was handled. These files could be subsequently accessed by project staff to review the data or by other programs in the project data system to calculate results to which the data contributed. Features were included in the K3ALQ program to estimate optimum aliquot weights for a specific purpose, to repeat measurements or operations in the event of spills or other mishaps, to test balance readings to determine whether they were within an expected range, and to guard against assigning the same identification number to more than one aliquot.

Illustrated below is the sequence of operations that was typically involved in aliquoting the dissolver solution samples for a particular segment.

When the set of dissolver samples was received at the Analytical Hot Cell, each sample vial was inspected for signs of breakage or mishandling, and its identity was checked for conformance to transfer documents that accompanied the samples from the DDS cell. Next, at a work station outside the hot cell, the analysts assembled materials and supplies appropriate to the types of aliquots that would be taken from each sample and labeled the assembled containers, etc., according to instructions in the written procedure (Document C-0030-0290 of Attachment 1) that directed the aliquoting operations. The supplies included a clean, new, weight buret (polyethylene wash bottle of 2-oz. capacity) for each solution sample and containers into which individual aliquots from each sample would be dispensed. The number and type of containers required for each solution sample were specified in the procedure according to the source code in the sample identification (see Section IV.A). Each DS sample or UB sample from a blank dissolution required a single 30-mL beaker that contained a previously prepared MSID spike containing 1 mg of NBS-993 (see Section B.1.a. of Appendix D for a description of the NBS 993 spikes). Each UB sample from a fuel-bearing segment required one such NBS 993 spike beaker; one

clean, new, 30-mL beaker; one clean, new liquid-scintillation-counting (LSC) vial (polyethylene); and one 50-mL polypropylene centrifuge cone. Each UB sample from a plenum segment required a single 30-mL beaker that contained a double MSID spike consisting of 20 μg of NBS 993 uranium and 100 μg ^{230}Th . Finally, each SB sample required only one new, clean 30-mL beaker. Each container was labeled with the appropriate sample identification and an analysis code corresponding to the type of aliquot it was to contain, e.g., UU, U5, U8, CS, or ZR (see Table 4 for a listing of analysis codes and definitions).

Having assembled the supplies required for aliquoting the sample at hand, the analyst called up the K3ALQ program on the computer terminal at the hot-cell work station. After prompting the analyst to enter his or her identity, the program asked for the identification of the segment for which samples would be processed. Based on this entry, the program determined whether the segment corresponded to a plenum segment (segment number 00), a fuel-bearing segment (number between 01 and 17), or a dummy segment (number between 91 and 99). It then checked that an appropriate DDSIF was available and opened the data files that contained information on the NBS 993 spikes and/or NBS 993/Th double spikes, as appropriate. It also determined whether samples from segments of the specified rod had been processed earlier, as evidenced by the existence of data files on samples and aliquots related to the rod of interest. If the sample and aliquot data files were not found, the program created new ones; if files existed, the program opened them to permit data retrieval and storage as required.

When the availability of all needed data files was assured, the program asked the analyst whether the samples to be processed were part of the usual set of samples for the type of segment that was specified or were being aliquoted for some special purpose. Subsequent operation of the program was virtually identical in either case. However, for the usual samples, the program anticipated the sample identifications and assigned aliquot identification numbers, whereas, for the special case samples, the analyst had to enter this information. Since special samples were rare during the EOL campaign, we describe only the usual case.

The program next prompted the analyst to select a specific sample from the expected sample set and to weigh the unopened sample vial on the remotely operated analytical balance (0.1-mg resolution) that was interfaced to the computer system. [This balance was calibrated and tested each working day by means of the balance performance program, BALQA, described in Section III.C.2.c.(1)] The analyst was given the option of skipping the specified sample and going on to the next one in the set. The program read the balance, computed the vial weight, and compared the weight with the full-bottle weight recorded for the specified sample in the DDSIF, or in the latest corresponding record of the sample information file if the sample had previously been processed through the K3ALQ program. If the measured vial weight agreed within 0.005 g of the reference weight from the appropriate data file, the program accepted the sample identity, recorded the apparent weight difference in the sample information file, and proceeded to the next operation. If the apparent weight exceeded the reference weight by more than 0.005 g, the analyst was directed to resolve the discrepancy, and the program proceeded to the next sample in the set. If the measured weight was smaller than the reference weight by more than 0.005 g, the analyst was directed to verify the sample's identity and was asked whether a correction for evaporation of the solution in the vial should be applied. Owing to

radiolytic decomposition of the dissolver solution samples and the vented-cap design of the sample bottles (see Section III.A.4.d), most of the solution samples processed during the EOL campaign lost weight before being aliquoted, and evaporation corrections were made. These corrections usually corresponded to a loss of about 20 mg ($\sim 0.06\%$ of the solution weight) per day. If so directed, the program calculated and stored an appropriate correction factor for relating the current and original concentrations in the sample solution, assuming the weight loss was due solely to vapor losses from the solution.

After the unopened sample vial was weighed and its weight was accepted, the analyst was prompted to open the vial, select and fill the correspondingly labeled weight buret with sample solution, and weigh the full buret. In the course of following these prompts, the analyst transferred the weight buret and containers assembled for the sample at hand into the hot cell. Only one weight buret and one set of containers were brought into the cell at a particular time to minimize potential mixup or cross-contamination of aliquots from the different samples. The program read the balance and stored the full-buret weight for use in subsequent calculation of the aliquot weight. It then prompted the analyst to select the labeled container corresponding to the type of aliquot that would be dispensed from the weight buret, displaying an appropriate description of the container as part of the prompt message.

If the specified container held an MSID spike, the analyst was prompted at this point to enter the spike identification number to the computer, and the program checked the appropriate spike files to verify that the specified spike had not been previously used for a different aliquot and that the identification was legitimate. When the aliquot was subsequently dispensed into the spike container, the aliquot identification was automatically logged into the appropriate record in the spike file, and pertinent data describing the spike were recorded in the file that contained information describing each aliquot associated with a given fuel rod.

After the analyst indicated, by terminal entry, that the appropriate container had been selected and the program verified and recorded the spike information as appropriate, the analyst was prompted to dispense the solution from the weight buret into the designated container. For aliquots that were destined for uranium analyses, the program computed, from the segment loading and solution weight data in the DDSIF file, the weight of solution required to provide approximately 1 mg of segment uranium in the aliquot and displayed this calculated weight on the terminal screen. This recommended weight of solution always was 10 g for DS samples and was restricted to the range of 1-10 g for UU and U8 aliquots. For U5 aliquots other than those from the DS samples, the minimum aliquot weight was set at 3 g to minimize relative errors in the measured weight value. For aliquots that were intended for fission-product measurements, the appropriate solution weight had to be deduced by the analyst from prior experience or trial and error. The aliquot weight in these cases was selected such that the aliquot's gamma activity did not exceed the count-rate capability of the gamma spectrometer system for an amount of solution not less than 0.1 g. In some cases, the sample solution had to be diluted by weight in order to achieve these criteria. Provision for carrying out such dilution under computer control was built into the K3ALQ program.

The analyst then dispensed the appropriate amount of solution from the weight buret into the designated container. To roughly gauge the weight of solution that was being dispensed, the container was placed on a second, low-resolution balance in the hot cell while the solution was being added. This rough weighing during dispensing eliminated many trial-and-error weighings of the buret, which would otherwise have been necessary to determine when the proper amount of solution had been dispensed. Since the weighings were performed with M/S manipulators, avoiding unnecessary repetitions represented a significant saving of effort.

After the aliquot was dispensed, the analyst was prompted to reweigh the buret on the computer-interfaced balance. The K3ALQ program calculated the weight of solution in the dispensed aliquot as the difference between the buret weights recorded before and after dispensing and also calculated a corresponding weight corrected for evaporation and dilution of the sample solution, as appropriate. Both calculated weights were stored in the data file containing aliquot information.

The program then proceeded to dispensing of the next aliquot from the sample being processed, and repeated the operations starting with filling of the weight buret with sample solution. When all aliquots from the given sample were dispensed, the analyst was prompted to close the sample vial and weigh it. This weight was stored in the sample information file for use as the sample reference weight if the sample required further aliquoting at a future time. The program then proceeded to direct the aliquoting of each additional sample appropriate to the specified segment type, repeating all of the above steps. The dispensed aliquots were submitted for further processing as discussed in Section IV.F.1.c.

Examples of the data files in which sample information and aliquot information were stored by the K3ALQ program are shown in Figures 18 and 19. In all, more than 700 solution samples were processed through the K3ALQ program during the EOL campaign, producing approximately 1600 separate aliquots for analysis. The value of the project computer system in handling this large volume of data that had to be acquired, stored, and processed

Sample I.D.	Empty Wt. of Vial, g	Vial Wt. Before Aliqtg., g	Wt. Diff., g	Vial Wt. After Aliqtg., g	Evap. Corr. Factor	Date	Analyst
B-00-UB-1	75.6462	105.1222	-4.512	93.5858	1.00000	23-AUG-84	RP,GK,M
B-00-UB-4	75.6672	105.4176	-0.056	93.6363	1.00191	23-AUG-84	RP,GK,M
B-02-DS-2	76.0130	107.1603	-0.014	96.9358	1.00044	23-AUG-84	RP,EK,M
B-02-UB-1	75.4660	108.6903	-0.028	105.8549	1.00083	23-AUG-84	RP,EK,M
B-02-UB-4	76.2390	111.2351	-0.059	108.7101	1.00168	24-AUG-84	EK,RP,M
B-02-SB-1	75.3010	115.1987	-0.091	114.0411	1.00229	27-AUG-84	EK,RP,
B-02-SB-4	76.1290	112.4249	-0.106	111.3476	1.00292	27-AUG-84	EK,RP,
B-03-DS-2	76.8593	113.3158	-0.019	102.6483	1.00053	27-AUG-84	EK,RP,M
B-03-SB-1	76.8583	110.6512	-0.027	109.4831	1.00079	27-AUG-84	EK,RP,M
B-03-SB-4	76.7913	107.1936	-0.029	106.0206	1.00097	27-AUG-84	EK,RP,M
B-03-UB-1	76.9863	112.9704	-0.042	110.6846	1.00116	27-AUG-84	EK,RP,M
B-03-UB-4	76.7768	111.8023	-0.040	108.1873	1.00113	27-AUG-84	EK,RP,M

Fig. 18. Example of Data File Containing Information on Samples Aliquoted during Analytical Operations. (Only a portion of the file for Rod B is shown.)

Aliquot Identification	Raw Wt., g	Dilution Factor	Corrected Wt., g	Spike I.D.	Wt. of NBS 993 in Spike, g	Wt. of Th-230 in Spike, g	Date	Analyst
B-00-UB-1-TH-1	11.1972	1.0000E+00	1.1197E+01	TH-109	2.01435E-05	1.10019E-04	23-AUG-84	RP, GK, M
B-00-UB-4-TH-1	12.3423	1.0000E+00	1.2366E+01	TH-110	2.04037E-05	1.09736E-04	23-AUG-84	RP, GK, M
B-02-DS-2-US-1	10.1433	1.0000E+00	1.0148E+01	93C-019	1.01821E-03	0.00000E-01	23-AUG-84	RP, EK, M
B-02-UB-1-UU-1	0.9854	1.0000E+00	9.8627E-01	NONE	0.00000E-01	0.00000E-01	23-AUG-84	RP, EK, M
B-02-UB-1-US-1	1.0522	1.0000E+00	1.0531E+00	93C-021	9.98321E-04	0.00000E-01	23-AUG-84	RP, EK, M
B-02-UB-1-DL-1	0.4351	4.2648E-02	4.3546E-01	NONE	0.00000E-01	0.00000E-01	23-AUG-84	RP, EK, M
B-02-UB-1-CS-1	0.5475	4.2648E-02	2.3350E-02	NONE	0.00000E-01	0.00000E-01	23-AUG-84	RP, EK, M
B-02-UB-1-ZR-1	2.0127	4.2648E-02	8.5838E-02	NONE	0.00000E-01	0.00000E-01	23-AUG-84	RP, EK, M
B-02-UB-4-UU-1	0.9863	1.0000E+00	9.6793E-01	NONE	0.00000E-01	0.00000E-01	24-AUG-84	EK, RP, M
B-02-UB-4-US-1	0.9973	1.0000E+00	9.9898E-01	93C-022	9.93081E-04	0.00000E-01	24-AUG-84	EK, RP, M
B-02-UB-4-DL-1	0.5165	5.7189E-02	5.1737E-01	NONE	0.00000E-01	0.00000E-01	24-AUG-84	EK, RP, M
B-02-UB-4-CS-1	0.4640	5.7189E-02	2.6536E-02	NONE	0.00000E-01	0.00000E-01	24-AUG-84	EK, RP, M
B-02-UB-4-ZR-1	1.8590	5.7189E-02	1.0631E-01	NONE	0.00000E-01	0.00000E-01	24-AUG-84	EK, RP, M
B-02-SB-1-UB-1	1.0245	1.0000E+00	1.0268E+00	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-02-SB-4-UB-1	1.0002	1.0000E+00	1.0091E+00	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-DS-2-US-1	10.0937	1.0000E+00	1.0099E+01	93C-023	9.98515E-04	0.00000E-01	27-AUG-84	EK, RP, M
B-03-SB-1-UB-1	1.0272	1.0000E+00	1.0280E+00	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-SB-4-UB-1	1.0087	1.0000E+00	1.0098E+00	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-1-UU-1	1.0347	1.0000E+00	1.0359E+00	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-1-US-1	1.0069	1.0000E+00	1.0081E+00	93C-024	9.95000E-04	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-1-DL-1	0.1917	2.3843E-02	1.9192E-01	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-1-CS-1	0.2499	2.3843E-02	5.9584E-03	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-1-ZR-1	1.1583	2.3843E-02	2.7427E-02	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-4-UU-1	1.0167	1.0000E+00	1.0178E+00	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-4-US-1	1.0476	1.0000E+00	1.0488E+00	93C-025	9.98558E-04	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-4-DL-1	0.2072	2.2815E-02	2.0743E-01	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-4-DL-2	0.2184	2.3034E-02	2.1059E-01	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-4-CS-1	0.2992	2.3034E-02	6.8916E-03	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M
B-03-UB-4-ZR-1	1.1363	2.3034E-02	2.6174E-02	NONE	0.00000E-01	0.00000E-01	27-AUG-84	EK, RP, M

Fig. 19. Example of Data File Containing Information on Aliquots Produced during Analytical Operations. (Only a portion of the file for Rod B is shown.)

is obvious. Computerization of the aliquoting procedure had the added advantage of reducing the demanding operations associated with hot-cell preparation of samples for high-precision analysis to a more-or-less routine state.

c. Preparation of Aliquots for Analysis

(1) Aliquots for Uranium Isotope Ratio Measurements

Uranium in each aliquot that was prepared for measurement of uranium isotope ratios had to be isotopically equilibrated, isolated, and purified prior to the isotopic measurements by thermal ionization mass spectrometry (TIMS) or APHA. Two different procedures were employed to accomplish these objectives. One of these procedures was used for aliquots of solution derived from fuel-bearing segments, for which only the uranium in each aliquot was of interest. The other procedure was used for aliquots of solution from the plenum dissolutions; in this case, both thorium and uranium in each aliquot had to be recovered for mass spectrometric analysis, and large amounts of radioactive and chemical impurities associated with dissolution of the plenum hardware had to be separated.

In both cases, isotopic equilibration was accomplished by taking each aliquot to gentle dryness several times from concentrated nitric acid.

This step also removed fluoride from each aliquot. Since subsequent measurements involved only determination of ratios of individual isotopes, quantitative recovery of the uranium in each aliquot during the separation and purification steps that followed this equilibration was not necessary. This feature of the ratio measurements greatly facilitated hot-cell operations involving these aliquots and is among the advantages associated with the isotope dilution technique for chemical assays.

For aliquots in which thorium was not of interest, (i.e., those from fuel-bearing segments), uranium in each aliquot was separated from thorium and most fission products by mixing a dilute nitric acid solution of salts from the dried aliquot with acid-deficient aluminum nitrate solution and extracting the uranium into methyl ethyl ketone (hexone). The extraction was performed inside the Analytical Hot Cell according to the procedure in Document C-0030-0291 of Attachment 1. The hexone layer containing the uranium from each aliquot was then transferred from the hot cell to the hood train in the uranium purification laboratory. There, uranium in the hexone layer was stripped by extraction into 1M HNO_3 and was purified by ion exchange according to the procedure in Document C-0030-0293 of Attachment 1.

In this procedure, the nitric acid solution containing the uranium was first taken to dryness and the residue redissolved in 0.1N H_2SO_4 . Then, the sample was loaded onto a previously conditioned anion exchange column. Iron was eluted from the column with 0.1N H_2SO_4 ; thorium, aluminum, sulfate ion, and nickel were eluted with 9M HCl ; and finally, uranium was stripped from the column with 0.1N HCl . The uranium in the eluate was converted to the nitrate salt and flamed to red heat to destroy organic matter, which could interfere with the mass spectrometric measurements. The resulting uranium oxide was dissolved in concentrated nitric acid and, if appropriate, a small aliquot (usually containing 1 to 2 μg U) was electrodeposited onto a planchet for APHA determination of the $^{232}\text{U}/^{233}\text{U}$ ratio. The remaining solution was transferred to a new, clean, 10-mL Pyrex beaker and evaporated to gentle dryness. The beaker was then submitted for mass spectrometric analysis of the uranium.

Uranium and thorium in the spiked aliquots of UB solution from the plenum dissolutions were isolated and purified according to the procedure in Document C-0030-0292 of Attachment 1. In this procedure, the salts obtained from drying the aliquot during the isotopic-equilibration step were dissolved in 8N HNO_3 , and the resulting solution was loaded onto a previously conditioned anion-exchange column inside the hot cell. The major fraction of radioactivity (from ^{60}Co , ^{137}Cs , ^{134}Cs , and ^{95}Zr) and chemical impurities such as Fe, Al, Ni, Cr, and Sn were washed through the column with 8N HNO_3 . Uranium and thorium were retained on the column. The anion-exchange column was then transferred from the hot cell to the hood train in the uranium purification laboratory where the thorium was eluted with 9M HCl and the uranium was eluted with 0.1M HCl . The separated uranium and thorium fractions were converted to the corresponding nitrate salts and submitted for mass spectrometric analysis.

(2) Aliquots for Fission Product Determinations

Solution aliquots that were intended for use in determining the concentrations of fission products (^{137}Cs , ^{144}Ce , and ^{95}Zr) in the UBT solution from each segment had to be brought to a condition that satisfied count-rate and geometry requirements for reliable measurements with the gamma spectrometer

system. In the case of aliquots designated for counting of ^{137}Cs and ^{144}Ce (i.e., aliquots with the CS analysis code), count-rate requirements were met by selecting the aliquot weight as the aliquot was dispensed (see Section IV.F.1.b). Geometry of each CS aliquot was adjusted to a predetermined configuration by adding 1.0N HNO_3 to the LSC vial that contained a given aliquot to bring the total volume of solution in the vial to 10 mL. To ensure maximum consistency in the counting geometries between samples and standards, all the polyethylene LSC vials that were used for preparing aliquots for gamma counting during the EOL campaign were selected from a single manufacturer's lot of vials. This precaution was taken after we found, during pre-EOL characterization work, that vials from different lots showed variations in their diameter up to several millimeters. Since our counting was carried out at a 40-cm distance from sample to detector, these variations could have introduced significant relative changes in the effective sample-to-detector distance for different samples and represented a potential source of random or systematic error in the measurements. About 2000 vials from a single lot, sufficient to accommodate the needs of the entire EOL campaign, were procured before the EOL work was begun. Measurements on vials selected at random from this supply showed that their diameters were uniform to within 0.5 mm, which would introduce negligible errors in the counting geometry.

The major fraction of cesium (^{134}Cs and ^{137}Cs) activity was removed from aliquots that were intended for determination of the ^{95}Zr concentration of the sample solution (aliquots having the ZR analysis code). This treatment reduced the total count rate obtained when the gamma spectrum of the aliquot was acquired and was applied so that larger solution aliquots could be used for the ^{95}Zr measurements, providing enhanced sensitivity for this nuclide. Because the cesium activity represented about 80% of the total radioactivity in the dissolver solutions, its removal permitted ZR aliquot weights that were up to five times larger than the corresponding CS aliquot weights.

Separation of the cesium activity in each ZR aliquot was carried out according to the procedure in Document C-0030-0298 of Attachment 1. The ZR aliquots had been dispensed into 50-mL polypropylene centrifuge tubes (see Section IV.F.1.b), which also contained Ce^{3+} and Ce^{4+} carriers added prior to dispensing the aliquot. In each centrifuge cone, $\text{Ce}(\text{OH})_x$ was precipitated from the solution by addition of ammonium hydroxide. In addition, ^{95}Zr and ^{144}Ce were quantitatively carried with the precipitate, along with U, Th, and several other fission products (notably ^{154}Eu) that were not of direct interest. Cesium activity remained with the supernatant. The supernatant was removed, and the precipitate was resuspension-washed with ammonium hydroxide solution. The washed precipitate was next dissolved in nitric acid, and the resulting solution was transferred into a clean, new, LSC vial. The solution in the vial was brought to the standard 10-mL volume by addition of 1.0M HNO_3 , and the vial was submitted for gamma counting. Corrections for small losses of the precipitate, which sometimes occurred during the washing and post-dissolution transfer steps in this procedure, were routinely made in subsequent calculation of the ^{95}Zr concentration of the aliquoted solution by using the apparent difference between ^{144}Ce concentrations determined for the untreated (CS) and treated (ZR) aliquots as a measure of the chemical yield from the separation process (see Section C of Appendix E for details of this calculation).

d. Measurement of Uranium Isotope Ratios

Ratios of each uranium isotope to ^{233}U were determined for each unspiked (analysis code = UU) aliquot of UBT solution from each fuel-bearing segment so that the segment uranium isotopic composition could be calculated. For isotopes other than ^{232}U , the ratios were directly measured by TIMS. Because of a potential isobaric interference by ^{232}Th in the TIMS measurement of ^{232}U , the ratio of the alpha-particle emission rate from the ^{232}U nuclide in the segment to the combined rate from ^{233}U plus ^{234}U was measured by APHA and was used together with the $^{234}\text{U}/^{233}\text{U}$ ratio from TIMS to compute the required $^{232}\text{U}/^{233}\text{U}$ value. The ratio of ^{233}U to the major spike isotope (i.e., ^{238}U for NBS 950a or ^{235}U for NBS 993) in samples prepared for MSID uranium assays was also measured by TIMS for each sample. Operations involved in the TIMS and APHA measurements are described immediately below; calculations performed with the ratios to obtain the uranium isotopic compositions and MSID assay results are described in Appendices B and D, respectively.

(1) Uranium Isotope Ratio Measurements by Thermal Ionization Mass Spectrometry

In TIMS, ions are produced at the surface of an electrically heated filament in the mass spectrometer ion source. A small amount of a sample containing the element of interest may be deposited directly on the ionizing filament (the so-called single-filament configuration) where the sample is evaporated and ionized simultaneously, or the sample may be loaded on and evaporated from a second filament, which is heated independently of the ionizing filament (the multiple-filament configuration). In either case, the observed ratio of ion currents corresponding to a ratio of isotopes in the sample may differ from the true ratio of isotopes, largely as the result of isotopic fractionation associated with preferential evaporation of lighter isotopic species. The conventional TIMS technique uses known mixtures of individual isotopes to empirically determine corrections for the bias errors associated with this fractionation. For this approach to be successful, a number of experimental variables must be held constant from run to run so that corrections determined from the standards will apply equally well to unknown samples analyzed under corresponding conditions. Among these variables are (1) the chemical form of the loaded sample, (2) the quantity of sample deposited on the filament, (3) the type of filament materials used, and (4) instrumental operating conditions during the course of each analysis.

For the isotope ratio measurements, two different approaches to the evaluation of fractionation corrections were used. One of these was based on the conventional method for making such corrections: a fractionation factor was determined by comparing the observed and certified ratios for a series of NBS uranium isotopic SRMs, and this factor was applied to the ratios measured for samples to correct for fractionation errors. The second approach was a unique, internal-standard procedure¹⁰ that was developed at ANL for the POB effort. In this procedure, a standard mixture of isotopes, different from the major isotopes in the sample, is added to the filament and, from the observed ratio of the internal-standard isotopes, a correction factor applicable to each individual analysis is determined. Because the quantity of internal standard added to the filament can be calculated from the observed isotopy of the standard/sample mixture, it is not necessary to precisely measure the quantity of internal standard that was added. However, because this calculation requires that the isotopic composition of the sample, as well as that of the internal standard, be known, its

application to isotopic composition determinations requires that each sample be analyzed twice, once with and once without added internal standard. The second analysis of each POB isotopic sample could not be justified by the relatively modest improvements in precision that would accrue to the isotopic abundance results if the internal-standard procedure were used. In contrast, owing to the availability of isotopic data for the segment and spike uranium in each MSID assay sample, the internal-standard approach could be used for these samples with virtually no added effort beyond programming the needed calculations. The internal-standard approach was, therefore, employed in measuring the $^{233}\text{U}/^{235}\text{U}$ ratio for samples from the SBT and the $^{233}\text{U}/^{235}\text{U}$ ratio in the UBT solution aliquots spiked with NBS 993. Spiked aliquots of the DS solution samples were not analyzed with the internal standard because the $^{233}\text{U}/^{235}\text{U}$ ratios in these samples were very small and their measurement could not be improved by reducing proportional error contributions.

Fractionation corrections were automatically applied to each measured ratio by the software associated with our fully automated VG-54R mass spectrometer [see Sections III.B.3 and III.C.2.b.(1) for descriptions of the instrument and software]. For samples run in the conventional way, the correction was evaluated from a running average of the ten most-recent analyses of NBS uranium isotopic standards from the series U-010 through U-930. In processing the data from a given sample run, the program searched a disk file of results with standards and computed the needed correction factors from the appropriate stored data. For the internal-standard runs, a mixture of ^{234}U and ^{236}U , which was calibrated against NBS U-500, was employed; this calibration was carried out by analyzing the $^{234}\text{U}/^{236}\text{U}$ mixture with U-500 as an internal-standard spike. Results of eight calibration measurements are given in Table 7, which illustrate the extremely high precision achievable for isotope ratio

Table 7. Results of Measurements from Calibration of $^{234}\text{U}/^{236}\text{U}$ Internal Standard

Run No.	$^{234}\text{U}/^{236}\text{U}$ Atom Ratio ^a
1	0.905687
2	0.905660
3	0.905666
4	0.905674
5	0.905669
6	0.905673
7	0.905674
8	<u>0.905670</u>
Average:	0.905672
Std. Deviation:	0.000008

^aDetermined with NBS U-500 as internal standard for each analysis.

determinations using the internal-standard procedure for fractionation corrections (% RSD of 0.0008% for the 8 runs). Abundances of minor isotopes in the internal-standard mixture are shown in Table 8.

Table 8. Abundances of Minor Isotopes in $^{234}\text{U}/^{236}\text{U}$ Internal Standard

Nominal Mass of Isotope	Atom Percent Abundance ^a
233	0.00521 $\pm$ 0.0001
235	0.00016 $\pm$ 0.0002
238	0.00007 $\pm$ 0.0001

^aNormalized to $^{234}\text{U}/^{236}\text{U} = 0.90567$.

To save time in analyzing EOL samples, we averaged fewer ratios during a given run than were averaged for the internal-standard calibrations; the TIMS instrument was programmed to continue each EOL run until the internal precision (i.e., the standard deviation of the mean of the individual ratios measured during a given run) reached 0.006%, relative. The % RSD estimated from analysis of duplicate SBT samples during the EOL campaign was 0.0062% (cf. Figure D-2 of Appendix D). This demonstrated that, with the internal-standard approach, a between-run (i.e., external) precision equal to the internal precision of the measurements could be achieved.

Uranium samples were received in the TIMS laboratory in the form of dried nitrate salt in 10-mL Pyrex beakers, which contained 1 to 2 mg of U. Uranium isotopic standards were kept in the laboratory as solutions, 1.0 mg U per milliliter in 0.8M HNO_3 , stored in Teflon bottles. Samples were loaded for analysis according to the written procedure in Document C-0030-0295 of Attachment 1. The nitrate salt was first dissolved in 0.8M HNO_3 to provide a concentration similar to that of the standards. The loading procedure was applied in identical fashion to samples and standards.

An aliquot of the uranium solution containing $1.0 \pm 0.2 \mu\text{g}$ of U was deposited on a tantalum mass-spectrometer filament, using a micro-syringe fitted with a disposable Teflon tip. For samples designated for MSID assay, a 1- μL aliquot ($\sim 1 \mu\text{g}$ U) of the ANL $^{234}\text{U}/^{236}\text{U}$ internal-standard solution was added to the sample drop on the filament. The solution was then evaporated to dryness, and the uranium was oxidized by passing a current through the filament. A programmable sample drier controlled the procedure. The drying sequence included heating the filament at 0.5 A for 10 min, followed by 1-min intervals at 1.0, 1.5, and 1.8 A.

The loaded filament was next incorporated, together with a second, blank, tantalum sample filament and a rhenium ionizing filament, into a triple-filament ionization assembly for subsequent insertion into the TIMS source. Up to 16 assemblies could be inserted into the instrument at one time and, after initiation of the measurement sequence, could be sequentially analyzed without operator intervention. In general, samples were loaded on filaments and inserted

into the mass spectrometer during the normal working day and were allowed to run overnight.

Operation of the TIMS instrument was directed by the written procedure in Document No. C-0030-0296 of Attachment 1. The operator entered appropriate sample identification and analysis information into the instrument's computer system and initiated the measurements. As each sample was run, results were stored automatically in redundant files on disk and on tape by the HP-9845 computer on the mass spectrometer. Each set of results was subsequently retrieved by the analyst through a special data-review program that permitted his or her evaluation of instrument performance during running of each sample and rejection of results for samples that did not run well. Data from each run judged acceptable by the analyst were transferred to the VAX computer and were summarized for computing isotopic or assay results for the segment to which they applied. An example of the data summary for samples from a specific segment is shown in Fig. 20. For each aliquot, the type of analysis (IS for internal standard or US for conventional), the date of analysis, and the values of individual measured ratios were recorded in the summary. Samples for which the analyst judged the run quality to be unsatisfactory were reloaded and rerun; if needed, specific samples were repurified or new aliquots were taken. Overall, reanalysis was called for during the EOL campaign in fewer than two of each 100 cases, attesting to the quality of both the sample preparation and the mass spectrometric procedure.

Uranium and thorium ratios in spiked aliquots of the UBT solutions from dissolved plenums were measured in a conventional manner with ANL's Teledyne mass spectrometer. Results of these measurements were manually entered into the VAX computer by means of a data-entry program that retrieved information on spike and aliquot weights and computed concentration values for the U and Th in the solution sample. These values were stored in a separate data file for access by project report-generator programs, which compiled data associated with each individual fuel rod. The data file of information on the LWBR fuel rod plenums is shown in Fig. 21.

(2) Uranium Isotope Ratio Measurements by Alpha Pulse Height Analysis

A portion of the uranium from each dissolved segment was analyzed by APHA to determine the $^{232}\text{U}/^{233}\text{U}$ isotope ratio. Samples designated for APHA were electrodeposited on stainless steel planchets following purification of the uranium by ion-exchange chromatography [see Section IV.F.1.c.(1)]. The samples were counted under vacuum ($\sim 25 \mu\text{m}$ pressure) using the EG&G ORTEC dual-port spectrometer described in Section III.B.5.

Operations related to obtaining and processing the alpha-emission spectrum from each sample were carried out according to the written procedure described in Document C-0030-0294 of Attachment 1. Manipulation and reduction of the alpha pulse data were performed with the aid of computer programs that were either purchased with the ORTEC system or developed at ANL. These programs utilized specific assumptions with regard to sample composition and intended use of the data and were tailored to the specific needs of the POB project. In particular, the data reduction routine assumed that alpha activity from nuclides other than ^{232}U , ^{233}U , ^{234}U , and ^{228}Th were negligible

B-01-DS-2-U5-1	3/5 Ratio	1.79284005E-02
B-01-DS-2-U5-1	Anal.Date	08-31-84
B-01-DS-2-U5-1	Anal.Type	US
B-01-SB-1-U8-1	3/8 Ratio	2.78124005E-01
B-01-SB-1-U8-1	Anal.Date	08-31-84
B-01-SB-1-U8-1	Anal.Type	IS
B-01-SB-4-U8-1	3/8 Ratio	2.78127998E-01
B-01-SB-4-U8-1	Anal.Date	08-31-84
B-01-SB-4-U8-1	Anal.Type	IS
B-01-UB-1-U5-1	3/5 Ratio	3.30832005E-01
B-01-UB-1-U5-1	Anal.Date	08-31-84
B-01-UB-1-U5-1	Anal.Type	IS
B-01-UB-1-UU-1	4/3 Ratio	7.21549010E-03
B-01-UB-1-UU-1	6/3 Ratio	1.22244994E-04
B-01-UB-1-UU-1	6/3 Ratio	1.64202004E-06
B-01-UB-1-UU-1	8/3 Ratio	2.90736003E-04
B-01-UB-1-UU-1	Anal.Date	08-31-84
B-01-UB-1-UU-1	Anal.Type	US
B-01-UB-4-U5-1	3/5 Ratio	3.44338000E-01
B-01-UB-4-U5-1	Anal.Date	08-31-84
B-01-UB-4-U5-1	Anal.Type	IS
B-01-UB-4-UU-1	4/3 Ratio	7.22143007E-03
B-01-UB-4-UU-1	6/3 Ratio	1.29877990E-04
B-01-UB-4-UU-1	6/3 Ratio	5.44846991E-06
B-01-UB-4-UU-1	8/3 Ratio	3.10789008E-04
B-01-UB-4-UU-1	Anal.Date	08-31-84
B-01-UB-4-UU-1	Anal.Type	US
B-02-DS-2-U5-1	3/5 Ratio	7.77801004E-01
B-02-DS-2-U5-1	Anal.Date	08-29-84
B-02-DS-2-U5-1	Anal.Type	US
B-02-SB-1-U8-1	3/8 Ratio	3.79078007E+00
B-02-SB-1-U8-1	Anal.Date	08-29-84
B-02-SB-1-U8-1	Anal.Type	IS
B-02-SB-4-U8-1	3/8 Ratio	3.79075003E+00
B-02-SB-4-U8-1	Anal.Date	08-29-84
B-02-SB-4-U8-1	Anal.Type	IS
B-02-UB-1-U5-1	3/5 Ratio	1.72869003E+00
B-02-UB-1-U5-1	Anal.Date	08-29-84
B-02-UB-1-U5-1	Anal.Type	IS
B-02-UB-1-UU-1	4/3 Ratio	5.68680018E-02
B-02-UB-1-UU-1	5/3 Ratio	6.81773992E-03
B-02-UB-1-UU-1	6/3 Ratio	8.65600014E-04
B-02-UB-1-UU-1	8/3 Ratio	2.93795997E-03
B-02-UB-1-UU-1	Anal.Date	08-29-84
B-02-UB-1-UU-1	Anal.Type	US
B-02-UB-4-U5-1	3/5 Ratio	1.64898002E+00
B-02-UB-4-U5-1	Anal.Date	08-29-84
B-02-UB-4-U5-1	Anal.Type	IS
B-02-UB-4-UU-1	4/3 Ratio	5.68640009E-02
B-02-UB-4-UU-1	5/3 Ratio	6.81660005E-03
B-02-UB-4-UU-1	6/3 Ratio	8.64194997E-04
B-02-UB-4-UU-1	8/3 Ratio	2.93944008E-03
B-02-UB-4-UU-1	Anal.Date	08-29-84
B-02-UB-4-UU-1	Anal.Type	US

Fig. 20. Summary Data File Containing Uranium Isotope Ratio Results from Thermal Ionization Mass Spectrometry. (Data for aliquots from segment B-01 are shown.)

relative to the activity from these four nuclides in the sample. Further, backgrounds were assumed to be small relative to the sample count rates so that background-subtraction contributions to uncertainty in the counting measurements could be ignored. No evidence to contradict these assumptions was found during the EOL work.

SUMMARY OF MASS SPECTROMETRIC RESULTS FOR PLENUM SEGMENTS

Aliquot Identification	Analyst	Date	Spike I.D.	Measured 233/235 U Ratio	Uranium Conc., g U per g Sol'n	Measured 232/230 Th Ratio	Thorium Conc., g Th per g Sol'n
A-00-UB-1-TH-1	ELC	8-JUN-84	TH-105	2.3480E-03	6.7497E-09	4.7480E-00	6.7748E-05
A-00-UB-4-TH-1	ELC	8-JUN-84	TH-105	2.3570E-03	6.4935E-09	5.0060E-00	6.7742E-05
B-00-UB-1-TH-1	ELC	28-AUG-84	TH-109	8.6000E-03	1.5443E-08	2.7330E-00	2.3889E-05
B-00-UB-4-TH-1	ELC	28-AUG-84	TH-110	9.3000E-03	1.5317E-08	3.0250E-00	2.3831E-05
C-00-UB-1-TH-1	ELC	24-SEP-84	TH-111	1.7140E-02	7.7407E-08	3.4750E-01	5.7319E-06
C-00-UB-4-TH-1	ELC	24-SEP-84	TH-112	1.1300E-02	5.8981E-08	2.7470E-01	4.7563E-06
D-00-UB-1-TH-1	ELC	26-OCT-84	TH-113	4.4130E-02	1.7585E-07	1.0443E-00	1.8900E-05
D-00-UB-4-TH-1	ELC	26-OCT-84	TH-114	5.3550E-02	1.7564E-07	1.2437E-00	1.8924E-05
E-00-UB-1-TH-1	ELC	16-NOV-84	TH-115	8.8100E-02	3.2879E-07	1.8860E-01	3.6259E-04
E-00-UB-4-TH-1	ELC	16-NOV-84	TH-116	1.0960E-01	3.2750E-07	2.3820E-01	3.6554E-04
F-00-UB-1-TH-2	ELC	10-DEC-84	TH-119	1.2360E-02	2.3887E-08	1.2530E-00	1.0546E-05
F-00-UB-4-TH-2	ELC	10-DEC-84	TH-120	1.1480E-02	2.3683E-08	1.1146E-00	1.0497E-05
G-00-UB-1-TH-1	ELC	17-JAN-85	TH-121	2.0615E-02	4.3761E-08	6.3728E-01	5.4264E-06
G-00-UB-4-TH-1	ELC	17-JAN-85	TH-122	2.2175E-02	4.3894E-08	6.4360E-01	5.4118E-06
H-00-UB-1-TH-1	ELC	29-JAN-85	TH-123	8.4020E-03	2.2641E-08	1.8109E-00	2.3295E-05
H-00-UB-4-TH-1	ELC	29-JAN-85	TH-124	1.1781E-02	2.1877E-08	2.5709E-00	2.3272E-05
I-00-UB-1-TH-1	ELC	4-MAR-85	TH-125	2.2110E-02	5.1361E-08	8.0700E-01	7.8837E-06
I-00-UB-4-TH-1	ELC	4-MAR-85	TH-126	2.2310E-02	4.5467E-08	8.7830E-01	7.8002E-06
J-00-UB-1-TH-1	ELC	19-MAR-85	TH-127	6.0530E-01	2.1200E-05	1.2309E-01	2.1568E-04
J-00-UB-4-TH-1	ELC	19-MAR-85	TH-128	5.9270E-01	2.1583E-06	1.2272E-01	2.2017E-04
K-00-UB-1-TH-1	ELC	11-APR-85	TH-130	3.1120E-01	6.2917E-07	1.1330E-02	1.1138E-03
K-00-UB-4-TH-1	ELC	11-APR-85	TH-131	2.8010E-01	6.2728E-07	1.0290E-02	1.1110E-03
L-00-UB-1-TH-1	ELC	21-MAY-85	TH-132	4.7150E-02	9.5895E-08	6.6160E-01	5.6334E-06
L-00-UB-4-TH-1	ELC	21-MAY-85	TH-133	4.8800E-02	9.5845E-08	6.3540E-01	5.6485E-06
M-00-UB-1-TH-1	ELC	26-JUN-85	TH-144	1.4638E-02	2.9283E-08	4.2850E-01	3.2904E-06
M-00-UB-4-TH-1	ELC	26-JUN-85	TH-145	1.2634E-02	2.8164E-08	3.9400E-01	3.2319E-06
N-00-UB-1-TH-1	ELC	18-JUL-85	TH-134	2.0470E-02	3.7861E-08	4.2810E-01	3.1723E-06
N-00-UB-4-TH-1	ELC	18-JUL-85	TH-135	1.7510E-02	3.7855E-08	3.6800E-01	3.1589E-06
O-00-UB-1-TH-1	ELC	6-AUG-85	TH-136	3.3600E-02	8.2730E-08	1.3550E-00	1.1391E-05
O-00-UB-4-TH-1	ELC	6-AUG-85	TH-137	3.2700E-02	6.2792E-08	1.2630E-00	1.1223E-05
P-00-UB-1-TH-1	ELC	4-SEP-85	TH-138	6.3200E-02	1.2402E-07	5.6360E-01	4.6523E-06
P-00-UB-4-TH-1	ELC	4-SEP-85	TH-139	6.3800E-02	1.2391E-07	5.6100E-01	4.6390E-06
Q-00-UB-1-TH-1	ELC	18-SEP-85	TH-140	1.5910E-02	3.1800E-08	3.3890E-01	2.4716E-06
Q-00-UB-4-TH-1	ELC	18-SEP-85	TH-141	1.5960E-02	3.1787E-08	3.3940E-01	2.4666E-06
R-00-UB-1-TH-1	ELC,ER	14-OCT-85	TH-142	8.4100E-03	1.3009E-08	3.6970E-01	2.7354E-06
R-00-UB-4-TH-1	ELC,ER	14-OCT-85	TH-143	8.0160E-03	1.2364E-08	3.6210E-01	2.7252E-06

Fig. 21. Data File Containing Results from Mass Spectrometric Measurements of Uranium and Thorium Isotope Ratios for Plenum Segments

During the APHA measurements, the stainless-steel planchet on which a uranium sample was deposited was inserted into the vacuum chamber of the alpha-pulse spectrometer counting port, the chamber was evacuated, and the alpha-pulse spectrum was acquired. Data were collected until at least 10,000 counts were accumulated in the smaller of the uranium peaks, or until 60,000 s had elapsed. Processing of the acquired spectrum was initiated by the analyst through commands to the spectrometer computer system and proceeded to completion without further operator intervention. The data-processing program first stored the raw spectrum in a file on disk. Then it created a new file by subtracting an appropriate background spectrum that had been acquired earlier and stored in a separate disk file. Separate regions of the net spectrum, each region containing a specific peak of interest, were integrated. These were (a) the region containing the characteristic alpha-decay energies of ^{234}U (4.77 and 4.72 MeV) and ^{233}U (4.82, 4.77, and 4.72 MeV), (b) the region containing the alpha-decay energies of ^{232}U (5.32, 5.26, and 5.13 MeV), and (c) the region that contained the 5.42 MeV peak of ^{228}Th .

The program then computed the counts in region (b) due to the 5.34 MeV peak of ^{228}Th [using the integrated counts from region (c) and branching information on alpha energies from ^{228}Th decay] and subtracted this contribution from the region (b) counts. For this calculation, branching factors of 72% for the ^{228}Th emission at 5.4 MeV and 28% for the peak at 5.3 MeV were presumed. Because ^{228}Th in the samples was removed during initial purification of the deposited uranium and accumulated solely as a product (daughter) of ^{232}U decay (half-life, about 70 y), this correction proved to be minor for all EOL samples, which were analyzed within a few days of being prepared. The program next computed the ratio of ^{232}U counts [i.e., those remaining in region (b)] to ^{233}U -plus- ^{234}U counts from region (a) and estimated the counting error associated with the ratio.

Finally, the results of these calculations were automatically stored in a file on disk, which also contained a summary of information that allowed identification of the raw spectra from which the data were obtained. An example of this alpha-counting summary data file is shown in Fig. 22. The $^{232}\text{U}/(^{233}\text{U} + ^{234}\text{U})$ count ratios stored in this file were subsequently used by project report-generator programs to compute the $^{232}\text{U}/^{233}\text{U}$ weight ratio for a given segment, as described in Section B.4 of Appendix B.

SAMPLE NO.	DET. I.D.	LIVE TIME SEC.	DATE OF BKG.	COUNTS AT 4.8 MEV	COUNTS AT 5.3 MEV	COUNTS AT 5.4 MEV	COUNT RATIO	% RSD RATIO	DATE OF COUNT	ANALYST
B-02-UB-4-UU-1	A	5638	28-AUG-84	328458	213858	719	0.502E-01	0.28	29-AUG-84	MWAE
B-02-UB-1-UU-1	B	5022	28-AUG-84	342198	222384	825	0.489E-01	0.27	29-AUG-84	MWAE
B-03-UB-1-UU-1	A	2021	28-AUG-84	128409	331227	419	2.578E+00	0.33	29-AUG-84	MW
B-03-UB-4-UU-1	B	1989	28-AUG-84	228327	587584	1419	2.571E+00	0.25	29-AUG-84	MW
B-01-UB-1-UU-1	A	5448	28-AUG-84	95923	24293	2	2.532E-01	0.72	30-AUG-84	MW
B-01-UB-4-UU-1	B	5397	28-AUG-84	97410	24520	5	2.517E-01	0.71	30-AUG-84	MW
B-04-UB-1-UU-1	A	2821	31-AUG-84	297541	1044217	1155	3.508E+00	0.21	04-SEP-84	MW
B-04-UB-4-UU-1	B	2803	31-AUG-84	230808	810452	1340	3.509E+00	0.24	04-SEP-84	MW
B-07-UB-1-UU-1	A	23350	31-AUG-84	220878	00977	75	4.118E-01	0.39	06-SEP-84	MWETK
B-07-UB-4-UU-1	B	23094	31-AUG-84	174850	71834	62	4.107E-01	0.44	06-SEP-84	K/W
B-05-UB-1-UU-1	A	6375	31-AUG-84	218897	599254	1346	2.735E+00	0.25	07-SEP-84	MW
B-05-UB-4-UU-1	B	6250	31-AUG-84	217383	594284	3772	2.727E+00	0.25	07-SEP-84	MW
B-06-UB-1-UU-1	A	2909	31-AUG-84	55348	44645	57	0.002E-01	0.64	07-SEP-84	MW
B-06-UB-4-UU-1	B	2893	31-AUG-84	66218	53000	158	0.005E-01	0.58	07-SEP-84	MW

Fig. 22. Example of Alpha-Counting Summary Data File

e. Automated Determination of Fission-Product Concentrations by Gamma Spectrometry

Concentrations of the fission products ^{137}Cs , ^{144}Ce , and ^{95}Zr in the UBT solution samples were determined through a fully automated procedure for gamma counting the weighed aliquots prepared as described in Section IV.F.1.c.(2). This procedure centered around the ORTEC 7050 Data Acquisition and Analysis System and ANL-built sample changer that were described in Section III.B.5, and the Alpha/Gamma Spectrometer software (GeLiGAM) mentioned in Section III.C.2.b.(2).

In using this system, the analyst positioned a series of counting samples and standards on the sample changer and entered specific information pertinent to each changer position into a file of control parameters. The analyst

then initiated an automatic procedure that brought the sample in each position to a location in front of the high-purity germanium detector, acquired and stored the gamma spectrum, and processed the data in the manner specified in the control-parameter file. Results of calculations performed with the data from each standard or sample were automatically written to an appropriate summary file for review by the analyst or access by project report-generator programs. Hardcopy printouts of the results of the spectrum analysis performed by the ORTEC GeLiGAM software were also generated by the gamma-counting programs and were individually reviewed by the analyst to ensure that peaks in each spectrum were consistent with the sample type, etc., specified in the control-parameter file.

The architecture of the automated gamma-counting procedure was modular by design to permit a single skeleton sequence of operations to be followed for every position of the sample changer, regardless of whether the position contained a sample or standard or was left empty. This skeleton sequence was controlled by a main operating program, written in command language, which executed any of a number of utility programs at specific points in the operating sequence. Each utility program performed a specific task such as assigning an identification number to the disk file in which a given spectrum would be stored, storing the spectrum, analyzing the spectrum, or calculating results pertinent to a given sample and writing the results to a specified data file. The identity of each utility program appropriate to the sample in any given changer position and values for parameters needed by the programs were selected by the main operating program from information coded in the control-parameter file. Many of the utility programs were provided by ORTEC in the GeLiGAM software package that was purchased with the spectrometer system; others were developed completely at ANL for specific application to the EOL samples from the POB project.

In the following subsections, we describe noteworthy features of the automated gamma-counting procedure and then present details of the data analysis that was applied in the case of each fission-product determination for the EOL samples.

(1) General Features of the Automated Gamma-Counting Programs

Operation of the automated gamma-counting programs centered around the control-parameter file, which was updated by the analyst each time samples were loaded into position on the automatic sample changer. An example of a completed control-parameter file is shown in Fig. 23. The data records in this file could be modified by the analyst through a data-entry program that used a "forms-driver" format. The program displayed the control-parameter file on the terminal screen as a data sheet to be filled out by the analyst. Data items needed only to be typed into predetermined fields under each heading in the file. The program automatically checked each entry to ensure that it was consistent with its required format (i.e., that it corresponded to a numeric value or an alphanumeric character string) and signaled the analyst if an inconsistency was detected. Individual entries could be modified or edited without altering any other parameters in the file.

The information in the control-parameter file for each occupied position of the sample changer is described below in subsections (a) through (e).

POS	SAMPLE ID	ANAL TYPE	PRES TYPE	PRESET VALUE	MAX CYC	TEST NUCLIDE	MAX RSD	INFORM FILE	CALIB FILE
1	081584BKG	BKG	LT	6000	1	NONE	50.0	S40BKG	SL40
2	137CSA1	CSSTD	LT	1900	3	CS-137	1.0	S40C7A	SL40
3	144CEA1	CESTD	LT	6400	3	CE-144	1.0	S40CEA	SL40
4	95ZRB1	ZRSTD	LT	6000	4	ZR-95	1.0	S40ZRB	SL40
5	B-01-UB-1-CS-1	POBCS	LT	1800	10	CS-137	0.5	S40POB	SL40
6	B-01-UB-4-CS-1	POBCS	LT	1800	10	CS-137	0.5	S40POB	SL40
7	B-01-UB-1-ZR-1	POBZR	LT	6000	10	ZR-95	2.0	S40POB	SL40
8	B-01-UB-4-ZR-1	POBZR	LT	6000	10	ZR-95	2.0	S40POB	SL40
9	137CSA1	CSSTD	LT	1800	3	CS-137	1.0	S40C7A	SL40
10	144CEA1	CESTD	LT	6400	3	CE-144	1.0	S40POB	SL40
11	95ZRB1	SKIP	LT	6000	10	ZR-95	2.0	S40POB	SL40
12	B-02-UB-1-CS-1	POBCS	LT	1800	10	CS-137	0.5	S40POB	SL40
13	B-02-UB-4-CS-1	POBCS	LT	1800	10	CS-137	0.5	S40POB	SL40
14	B-02-UB-1-ZR-1	SKIP	LT	6000	10	ZR-95	3.0	S40POB	SL40
15	R-09-UB-4-ZR-1	STOP	LT	6000	10	ZR-95	3.0	S40POB	SL40
16									
17									
18									
19									
20									
21									
22									
23									
24									
25									
26									

Fig. 23. Example of a Typical Control-Parameter File for Use in the Automated Gamma-Counting Procedure. (Positions 16-26 are empty.)

(a) Sample Identification

The sample in each changer position was assigned an identification number by the analyst based on the following three rules. One, if the sample corresponded to an empty or blank position for use in determining the current background spectrum, the sample was named with a six-digit number corresponding to the date of analysis followed by the letters "BKG." Two, if the changer position contained a working standard consisting of a solution of one of the fission products of interest, it was assigned the identification of the standard, which had to be consistent with one of the entries in a data file describing each standard that was approved for use in the project (see Fig. 24 for a listing of the information in this file). Three, if the changer position contained an aliquot of dissolver solution prepared for gamma-counting of fission products, it was assigned the same sample designation as the aliquot.

(b) Types of Data Analysis

Also included was a control parameter corresponding to a coded description of the specific operations that were to be executed in processing the data obtained from the gamma spectrum for the given sample. Ten different analysis types, described in Table 9, were recognized by the operating programs. Based on the analysis-type designation for a given sample, the main operating program selected appropriate utility programs to store the acquired spectrum in a data file named with the sample number and a characteristic suffix, to analyze the spectrum by integrating specific peaks and making corrections for potentially interfering peaks from nuclides that were not of direct

INFORMATION ON STANDARDS - FILE GAMSTDS.DAT

STANDARD I.D.	SOURCE	ANALYSIS DATE	ANAL TIME	ZONE	NUCLIDE	MICRO CI ACTIVITY
137CSA1	NBS4233B	29-AUG-79	00:01	EST	CS-137	1.8164E+01
137CSA2	NBS4233B	29-AUG-79	00:01	EST	CS-137	1.8164E+01
137CSA3	NBS4233B	29-AUG-79	00:01	EST	CS-137	1.8184E+01
134CSA1	AMERSHAM	01-JUN-83	12:00	GMT	CS-134	1.253E+01
134CSA2	AMERSHAM	01-JUN-82	12:00	GMT	CS-134	1.253E+01
134CSA3	AMERSHAM	01-JUN-82	12:00	GMT	CS-134	1.255E+01
144CEA1	AMERSHAM	09-MAR-83	12:00	GMT	CE-144	4.278E+01
144CEA2	AMERSHAM	09-MAR-83	12:00	GMT	CE-144	4.307E+01
144CEA3	AMERSHAM	09-MAR-83	12:00	GMT	CE-144	4.042E+01
154EUA1	AMERSHAM	01-JUN-82	12:00	GMT	EU-154	1.697E+01
154EUA2	AMERSHAM	01-JUN-82	12:00	GMT	EU-154	1.709E+01
154EUA3	AMERSHAM	01-JUN-82	12:00	GMT	EU-154	1.702E+01
95ZRA1	AMERSHAM	01-JUN-83	12:00	GMT	ZR-95	1.291E+01
95ZRA2	AMERSHAM	01-JUN-83	12:00	GMT	ZR-95	1.314E+01
95ZRA3	AMERSHAM	01-JUN-83	12:00	GMT	ZR-95	1.311E+01
95ZRA4	AMERSHAM	01-JUN-83	12:00	GMT	ZR-95	4.413E+01
95ZRB1	AMERSHAM	24-MAY-84	12:00	GMT	ZR-95	1.307E+02

Fig. 24. Data File Describing Working Standards
Used in Application of the Automated
Gamma-Counting Procedure

interest, to calculate results, and to write the results to designated data files. The analysis types included some (e.g., POBCS, POBZR, XXSTD) that were specific to samples anticipated in the EOL work, along with some (e.g., POBSL and POBPT) that could be used for general screening measurements or for special samples that were not anticipated prior to the campaign.

(c) Spectrum-Acquisition Features

Five separate parameters (preset type, preset value, maximum cycles, test nuclide, maximum RSD) were specified for each changer position to control acquisition of the sample's gamma spectrum. These permitted the analyst to take advantage of acquisition options provided by the GeLiGAM software and to utilize a feature of the ANL programs that optimized the time spent in counting any given sample.

The GeLiGAM programs acquire counts in the spectrum until some preset criterion is met, at which point acquisition is stopped and the acquired spectrum may be manipulated. The type of criterion [options include live time (LT), real time (RT), region-of-interest counts (RI), and net counts (NC)] and the value at which acquisition is to terminate are specified by the preset type and preset value (Fig. 23). For our EOL work, the LT preset type was used exclusively and was set to a relatively short time interval.

A spectrum can be acquired over a series of short intervals, each corresponding to the time required for meeting the preset criterion. At the end of each acquisition interval, the spectrum is analyzed by means of the GeLiGAM program specified according to the analysis-type parameter, and the results of this analysis are used to determine whether counting statistics achieved for a specific test nuclide in the sample were satisfactory. If the % RSD counting error associated with the test nuclide is smaller than a maximum acceptable value (the MAX RSD specified in the control-parameter file), acquisition is terminated and the data from the spectrum analysis are processed.

Table 9. Types of Analysis Available for Execution
by the Automated Gamma-Counting Programs

	Analysis Type	Description
1.	BKG	Store spectrum with suffix .BKG; analyze spectrum and store results for use in peaked-background correction calculations.
2,3,4.	XXSTD (XX = CS, CE, ZR)	Store spectrum with suffix .SPC; analyze spectrum and compare results with data for corresponding standard in file containing information on standards; write results to appropriate quality assurance file for measurements on specified nuclide.
5.	POBCS	Store spectrum with suffix .SPC; analyze spectrum and convert results for ^{137}Cs , ^{134}Cs , and ^{144}Ce to atoms-per-gram-of-solution concentrations; write results to summary data file for CS aliquots related to rod specified in sample identification.
6.	POBZR	Store spectrum from first cycle with suffix .CER, others with suffix .SPC; analyze spectra and convert results for ^{144}Ce and ^{95}Zr to atoms-per-gram-of-solution concentrations; write results to summary data file for ZR aliquots related to rod specified in sample identification.
7.	POBSL	Store spectrum with suffix .SPC; analyze spectrum as for solution samples and provide hardcopy output of results; no data written to summary files.
8.	POBPT	Store spectrum with suffix .SPC; analyze spectrum as for point source and provide hardcopy output of results; no data written to summary files.
9.	SKIP	Skip the sample in this position.
10.	STOP	Terminate execution of the automated procedure when this position is reached.

Otherwise, acquisition of counts in the spectrum is continued for another interval, and the testing cycle is repeated until either the desired counting statistics are achieved or a specified maximum number of cycles is reached. By keeping each acquisition interval relatively short, we could keep the total time required to achieve a desired level of precision for counting of the test nuclide in the sample to a minimum. At the same time, longer counts were available for samples that might require them so that time-consuming recounts on low-level samples could be avoided. The efficient use of available counting time that was provided by this feature of the automated gamma-counting procedure proved invaluable in

helping to maintain the rapid pace and high quality required by the EOL operations.

A special spectrum-acquisition feature was built into the procedure used for the POBZR analysis. Because the activity of ^{144}Ce in the dissolver solution samples was much higher than that of ^{95}Zr (the test nuclide for ZR aliquots), the 133.5 keV peak for ^{144}Ce in the acquired spectrum frequently overflowed the multichannel analyzer before sufficient ^{95}Zr counts were accumulated to meet the desired counting statistics. To permit both nuclides to be determined with acceptable precision from a single counting of the sample, the spectrum acquired during the first acquisition cycle was stored with a .CER suffix and was analyzed to provide the ^{144}Ce measurement. The preset value for each cycle was selected to provide adequate statistics for this measurement. Acquisition was then continued until terminated in the manner described earlier. The spectrum ultimately used for the ^{95}Zr determination was stored with the usual suffix (.SPC). If the spectrum from the first cycle satisfied the ^{95}Zr counting criterion, the same spectrum was stored and analyzed twice, once with the .CER suffix and once with the .SPC suffix.

Acquisition-control parameters required for achieving count-time optimization (the identity of the test nuclide, the maximum acceptable % RSD for its counting, and the maximum number of cycles) were specified for each sample position in the control-parameter file by the analyst. If the test nuclide were specified as "NONE" (cf. Fig. 23), only one cycle would be executed for the sample in that position.

(d) Spectrum-Analysis Features

Data-reduction routines available in the GeLiGAM software package were used for analyzing the spectral data acquired for each sample that was processed with the automated gamma-counting system. The GeLiGAM system is modular and works from (1) user-built libraries of isotopic nuclear data, (2) data files that inform the programs regarding user-selected options and parametric values (so-called INFORM files), and (3) calibration files created from GeLiGAM analysis of standards containing known nuclides at known concentration. The INFORM and calibration files specified for analyzing a particular spectrum are stored with the raw-spectrum data to diminish the possibility of error and to facilitate auditing. Details of the GeLiGAM system and its options are available in Reference 11.

Particular data-reduction routines for use with spectra acquired with our system were coded in the analysis-type control parameter. For each analysis involving determination of selected nuclides in a given sample (e.g., type POBCS, POBZR, or XXSTD), we used the routine called GAMMA3, which performs a three-step analysis of the spectrum. In the first step, the spectrum is analyzed on the basis of interference-free peaks corresponding to nuclides specified in the first of three library files. In our system, this library contained information on peaks from ^{137}Cs (661.6 keV), ^{144}Ce (133.5 keV), ^{134}Cs (604.7 and 795.8 keV), and ^{154}Eu (1004.8 and 1274.5 keV). Next, GAMMA3 computes the counts expected from these nuclides at peak energies other than those listed in the first library file. The second library file specifies the peaks for which counts are calculated based on the measured, interference-free peaks. Our second library contained information on the ^{154}Eu peaks at 723.3 and 756.9 keV, which interfered with determination of ^{95}Zr (peak energies at 724.2 and 756.7 keV).

The calculated counts at each peak region specified in the second library file are subtracted from the net counts for that region and added to the region background. Finally, interference-corrected peaks in the third library file (^{95}Zr and ^{95}Nb in our case) are analyzed, and results for each individual nuclide are written to an output file.

The GAMMA3 routine uses a specified calibration file to convert count rates at the given peak energies to activities of the corresponding nuclides and refers to a specified INFORM file to determine whether corrections for peaked backgrounds or radioactive decay of the nuclides are to be made. The INFORM file also includes values of parameters needed for such corrections, e.g., the reference time to which decay corrections should be made. Calibration and INFORM files used for processing spectra from EOL samples were set up prior to the campaign. The analyst selected the proper files based on the type of samples being analyzed and entered the file designation in the appropriate record of the control-parameter file as each sample was loaded for counting with the automated system.

The output of the GAMMA3 analysis includes values for the decay- and peaked-background-corrected activities of individual nuclides in the sample, as well as estimates of the counting errors associated with each activity value. Activities were given in units of microcuries present in the sample that was counted; errors were listed as the % RSD uncertainty based on counting statistics. Decay corrections were made to 12:00 (noon) on January 1, 1984, for nuclides in dissolver solution samples and to the certification date for nuclides in the working standards that were counted (cf. Fig. 24).

(e) Data Handling and Summary

Data in the output file generated by each GAMMA3 spectrum analysis were read and reformatted by other programs in the automated counting system. This provided data summaries that could be perused by the analyst for review of the operations or accessed by report-generator programs in performing calculations that involved the gamma-counting data. The programs that produced summary data files were of two types.

One type was part of the GeLiGAM software package. It produced a comprehensive summary report of the spectrum-analysis results, including the characteristic energies of peaks found in the spectrum, values for count-rate and peak-shape parameters associated with each peak, the identity of options selected in processing the spectrum, and activities of each nuclide identified from the spectrum. This summary report was printed in hardcopy at the gamma-counting station each time a spectrum was analyzed with GeLiGAM software, e.g., at the end of each acquisition cycle during the counting of a given sample. It was used by the analyst to verify that each spectrum was correctly processed.

The other type of data-summary program consisted of programs developed at ANL to compile data from related samples in a form that permitted the analyst to easily compare results from duplicate determinations or to otherwise judge the quality of measurements made with the automated system. Three such programs were routinely used during the EOL campaign.

One of these was called by the main operating program when the analysis involved one of the ^{137}Cs , ^{144}Ce , or ^{95}Zr working standards. This program (1) read the decay-corrected microcurie activity for the appropriate nuclide from the GAMMA3 output file, (2) retrieved information on the specified standard from the standards information file (Fig. 24), (3) calculated parameters that described the difference between the measured and theoretical values for the standard, and (4) wrote a record describing the results of the standard run to a quality-assurance summary file. Separate files were kept for each of the three fission-product nuclides for which standards were routinely analyzed. One of these files is illustrated in Fig. 25, which shows a portion of the quality-assurance summary file for runs with the ^{137}Cs standards. Corrections for bias between the measured and theoretical values for each standard were applied to results obtained for samples analyzed during the same time period. This action was taken by project report-generator programs that accessed the data in these quality-assurance summary files. Calculation of the correction for apparent counting bias is outlined in Section D of Appendix E.

STANDARD ANALYSIS QUALITY ASSURANCE TABLE

STANDARD RUN	ANALYSIS DATE	ANALYSIS TIME	EXPECTED MICRO CI	FOUND MICRO CI	COUNT RSD	PCT DIFF	ERROR SIGMA
137CSA1	10-AUG-84	16:08:31	1.8184E+01	1.8149E+01	0.3	0.1	0.2
137CSA2	10-AUG-84	16:41:29	1.8184E+01	1.8173E+01	0.3	0.1	0.2
137CSA3	10-AUG-84	17:14:27	1.8184E+01	1.8207E+01	0.3	0.1	0.4
137CSA1	24-AUG-84	17:11:02	1.8184E+01	1.8102E+01	0.3	0.3	1.0
137CSA1	28-AUG-84	16:46:16	1.8184E+01	1.8061E+01	0.3	0.6	1.7
137CSA1	29-AUG-84	16:44:30	1.8184E+01	1.8071E+01	0.3	0.6	1.5
137CSA1	30-AUG-84	17:06:28	1.8184E+01	1.8019E+01	0.3	0.8	2.4
137CSA1	04-SEP-84	17:05:31	1.8184E+01	1.8132E+01	0.3	0.2	0.6
137CSA1	06-SEP-84	16:59:15	1.8184E+01	1.8087E+01	0.3	0.4	1.3
137CSA1	07-SEP-84	16:38:11	1.8184E+01	1.8035E+01	0.3	0.7	2.1
137CSA1	20-SEP-84	17:08:32	1.8184E+01	1.8151E+01	0.3	0.1	0.2
137CSA1	21-SEP-84	17:17:07	1.8184E+01	1.8099E+01	0.3	0.4	1.1
137CSA1	24-SEP-84	17:03:30	1.8184E+01	1.8009E+01	0.3	0.9	2.5
137CSA1	25-SEP-84	16:33:17	1.8184E+01	1.8133E+01	0.3	0.2	0.6
137CSA1	27-SEP-84	18:06:58	1.8184E+01	1.8041E+01	0.3	0.7	2.0
137CSA1	28-SEP-84	18:18:45	1.8184E+01	1.8136E+01	0.3	0.2	0.5
137CSA1	01-OCT-84	18:00:00	1.8184E+01	1.8074E+01	0.3	0.5	1.5
137CSA1	04-OCT-84	15:10:31	1.8184E+01	1.8020E+01	0.3	0.8	2.3
137CSA1	05-OCT-84	17:30:54	1.8184E+01	1.8113E+01	0.3	0.3	0.8
137CSA1	08-OCT-84	15:30:52	1.8184E+01	1.8180E+01	0.3	0.1	0.3
137CSA1	10-OCT-84	13:49:27	1.8184E+01	1.8211E+01	0.3	0.3	0.8
137CSA1	12-OCT-84	16:55:33	1.8184E+01	1.8132E+01	0.3	0.2	0.6
137CSA1	24-OCT-84	16:42:23	1.8184E+01	1.8075E+01	0.3	0.5	1.4
137CSA1	26-OCT-84	16:54:36	1.8184E+01	1.8074E+01	0.3	0.5	1.5
137CSA1	30-OCT-84	17:47:48	1.8184E+01	1.8024E+01	0.3	0.8	2.3
137CSA1	31-OCT-84	18:00:25	1.8184E+01	1.8102E+01	0.3	0.6	0.8
137CSA1	02-NOV-84	17:38:50	1.8184E+01	1.8004E+01	0.3	0.9	2.6
137CSA1	05-NOV-84	15:26:37	1.8184E+01	1.8106E+01	0.3	0.3	1.0
137CSA1	08-NOV-84	17:34:01	1.8184E+01	1.8014E+01	0.3	0.8	2.4
137CSA1	14-NOV-84	18:18:02	1.8184E+01	1.8027E+01	0.3	0.8	2.2
137CSA1	16-NOV-84	14:48:41	1.8184E+01	1.8000E+01	0.3	0.9	2.7
137CSA1	19-NOV-84	18:10:22	1.8184E+01	1.8202E+01	0.3	0.2	0.6
137CSA1	20-NOV-84	18:51:51	1.8184E+01	1.8088E+01	0.3	0.4	1.2
137CSA1	21-NOV-84	17:53:16	1.8184E+01	1.8235E+01	0.3	0.4	1.2
137CSA1	26-NOV-84	18:18:44	1.8184E+01	1.8127E+01	0.3	0.2	0.6
137CSA1	27-NOV-84	18:20:38	1.8184E+01	1.7983E+01	0.3	1.0	2.9
137CSA1	28-NOV-84	17:18:49	1.8184E+01	1.8142E+01	0.3	0.1	0.4

Fig. 25. Portion of Quality-Assurance Summary File Containing Results of Measurements on ^{137}Cs Working Standards

The other two data-summary programs developed at ANL were used in treating data obtained from the counting of dissolver-solution samples to determine their fission-product content. Like the program for treating standards data, these two programs also read the microcurie activities for selected fission-product nuclides from the GAMMA3 output file but then converted the microcurie units to number of atoms and computed the concentration of each nuclide in the sample solution by dividing the number of atoms in the counted aliquot by the aliquot weight recorded when the aliquot was taken (see Section IV.F.1.b and Fig. 19). When ^{144}Ce was one of the nuclides of interest, the program also applied corrections for absorption of the ^{144}Ce gamma rays by Th and U in the sample solution. The resulting concentration data were summarized in a data file designated to contain either Cs and Ce (CS aliquots, POBCS analysis type) or Zr and Ce (ZR aliquots, POBZR analysis type) results for samples from a given fuel rod. Calculations involved in the units conversion and absorption corrections are described below.

(2) Computation of Fission-Product Concentrations in Dissolver-Solution Samples

The concentration, C_{FP} , of a particular fission product in a given dissolver-solution sample was obtained by dividing the number of atoms, N_{FP} , of the fission product in the solution aliquot that was counted by the weight, W_{ALQ} , of solution that the aliquot contained, i.e.,

$$C_{FP} = N_{FP}/W_{ALQ} \quad (\text{IV-7})$$

The value of W_{ALQ} for this calculation was retrieved by the appropriate data-summary program from the file of aliquoting data for samples from the rod to which the sample identification corresponded. The program read the corrected aliquot weight from the data record for the specified aliquot (see Fig. 19), thereby taking into account the evaporation and dilution corrections that were previously applied to the value (Section IV.F.1.b) and, hence, using a value that corresponded to the weight of as-sampled BT solution contained in the aliquot.

The value for N_{FP} was computed by the data-summary program from the microcurie activity, A_{FP} , of the specified fission product, as obtained from analysis of the aliquot's gamma spectrum in the GAMMA3 output file [see Section IV.F.1.e.(1).(e)]. Formulas for converting microcuries to atoms were derived for each fission-product of interest by applying the relationship

$$N_{FP}\lambda_{FP} = 3.7 \times 10^4 A_{FP} \quad (\text{IV-8})$$

where λ_{FP} is the decay constant (in units of s^{-1}) for the specified nuclide. Values for λ_{FP} were obtained from each fission-product half-life, T_{FP} , and the relationship

$$\lambda_{FP} = \ln 2 / T_{FP} \quad (IV-9)$$

Half-lives used in our calculations are listed in Table 10, together with conversion factors derived from the half-lives to convert microcuries to atoms. In the data-summary programs, N_{FP} was calculated by simply multiplying A_{FP} by the appropriate conversion factor.

Table 10. Half-Lives of Selected Fission Products and Factors for Converting Microcurie Units to Atoms

Fission Product	Half-Life	Conversion Factor, atoms per μCi
^{137}Cs	30.17 years	5.0828×10^{13}
^{134}Cs	2.062 years	3.4734×10^{12}
^{144}Ce	284.3 days	1.3112×10^{12}
^{95}Zr	64.02 days	2.9526×10^{11}

When ^{144}Ce was one of the fission products of interest in the counting sample, a correction was applied to account for absorption of the 133.5 keV gamma rays by Th and U in the sample solution. This correction was necessary because this photon energy falls on an absorption edge in the photon-absorption characteristic curves for Th and U and might be significantly attenuated by these actinide elements. In making this correction, we treated solution in the LSC vial that contained the counting sample as a cylindrical radiation source. Self-absorption of gamma rays leaving such a source may be described by the approximate expression¹²

$$N = N_0 \exp [-(8/3\pi)\mu\rho R] \quad (IV-10)$$

Here, N is the number of photons leaving the source, N_0 is the number of photons that would be emitted in the absence of self-absorption, μ is the mass absorption coefficient for photons of the specified energy, ρ is the density of the absorber in the radiation source, and R is the radius of the cylinder that the source represents. Values of μ for Th and U are available in Ref. 13 as a function of photon energy; the values for these elements are practically the same at 133.5 keV. A value for R was determined from the diameter of the LSC vials used for the gamma-counting samples. As discussed in Section IV.F.1.c.(2), these vials were selected to have a uniform diameter from vial to vial, yielding a single value of R to be applied to all EOL samples.

The density of the absorber in each counting sample was calculated as the mass of Th plus U in the sample divided by the standard volume (10 mL) to which solution in each vial was brought during its preparation for counting. The mass of Th plus U was, in turn, estimated from data available in the DDSIF file (Section IV.F.1.a), using the relationship

$$W_{Th,U} = (W_{OX}W_{ALQ}/W_{UBT})0.8788 \quad (IV-11)$$

Here, W_{OX} is the weight of oxide (ThO_2 plus UO_2) estimated to have been present in the segment from which the sample was derived, W_{ALQ} is the corrected weight of the UBT solution aliquot contained in the counting sample, W_{UBT} is the weight of solution in the UBT (presumed to contain the entire dissolved segment), and 0.8788 is the stoichiometric factor relating Th to ThO_2 . The value of this last factor changes negligibly if the stoichiometric factor for ^{233}U vs. $^{233}UO_2$ is used. Thus, Eq. IV-11 accounts, to a good approximation, for the total mass of Th and U in the counting sample, regardless of the distribution of U and Th in the oxide. The formula used in applying the self-absorption correction to each ^{144}Ce concentration that was measured is

$$^{Corr}C_{^{144}Ce} = ^{App}C_{^{144}Ce} \cdot \exp(0.37763W_{Th,U}) \quad (IV-12)$$

Here, the superscripts Corr and App indicate the corrected and apparent ^{144}Ce concentrations, respectively.

In general, because the aliquots taken for gamma counting were small, the self-absorption corrections increased the apparent ^{144}Ce concentrations by only 2% or less. Thus, the approximations involved in deriving the formula for applying these corrections introduced negligible error into the resulting concentration values.

Fission product concentrations, calculated in the manner outlined above, were written to summary data files by the data-summary programs. Each summary data file listed the date and time of analysis of the sample and % RSD values estimated for each concentration by the GeLiGAM programs. Two summary data files were produced for each LWBR fuel rod. One file contained results from the gamma counting of CS aliquots, as illustrated for Rod B in Fig. 26a; the other contained results from counting the corresponding ZR aliquots, as shown in Fig. 26b. In generating records in this latter file, our program calculated the ^{144}Ce concentration from analysis of the .CER spectrum from the indicated sample and the ^{95}Zr concentration was calculated from the corresponding .SPC spectrum.

Data in these summary files were subsequently accessed by project report-generator programs for computing the fission-product loading of each fuel-rod segment, as described in Section V.F and Appendix E.

2. Analysis of Gas Samples by Mass Spectrometry

Gas samples collected during puncture of each fuel rod's plenum or dissolution of each segment were analyzed by mass spectrometry to determine the gas composition. The isotopy of the Kr and Xe fission products in the plenum-gas samples was also determined; corresponding isotopic measurements on the off-gas samples from the dissolver were not performed because concentrations of Xe and Kr were too low to permit reliable measurements. Instrumentation for these analyses was described in Section III.B.4. Here, we present a brief description

CESIUM SAMPLE RESULT TABLE FOR ONE ROD ON NOON 01-JAN-84

SAMPLE I.D.	ANALYSIS DATE	ANALYSIS TIME	CS-134 ATM/G	CS-134 RSD	CS-137 ATM/G	CS-137 RSD	CE-144 ATM/G	CE-144 RSD
B-02-UB-1-CS-1	24-AUG-84	19:09:34	2.275E+15	0.5	9.303E+18	0.3	8.025E+15	0.4
B-07-UB-1-CS-1	06-SEP-84	19:27:42	8.289E+12	1.3	1.216E+15	0.3	5.935E+13	0.6
B-07-UB-4-CS-1	06-SEP-84	20:34:58	6.223E+12	1.2	1.210E+15	0.3	5.852E+13	0.5
B-05-UB-1-CS-1	06-SEP-84	21:43:23	8.543E+15	0.3	2.055E+17	0.3	9.244E+15	0.5
B-05-UB-4-CS-1	06-SEP-84	22:51:40	8.506E+15	0.4	2.034E+17	0.3	9.437E+15	0.7
B-06-UB-1-CS-1	06-SEP-84	23:59:13	2.187E+15	0.8	9.607E+18	0.3	3.665E+15	0.7
B-06-UB-4-CS-1	07-SEP-84	01:06:17	2.203E+15	0.8	9.684E+18	0.3	3.646E+15	0.9
B-02-UB-4-CS-1	24-AUG-84	19:36:32	2.281E+15	0.4	9.327E+18	0.2	6.074E+15	0.4
B-03-UB-1-CS-1	28-AUG-84	18:56:23	8.155E+15	0.4	1.925E+17	0.3	9.918E+15	0.6
B-01-UB-1-CS-1	28-AUG-84	23:13:17	1.727E+12	1.7	3.949E+14	0.3	5.064E+13	0.4
B-01-UB-4-CS-1	29-AUG-84	00:56:42	1.698E+12	1.3	3.960E+14	0.3	5.107E+13	0.3
B-03-UB-4-CS-1	28-AUG-84	19:34:40	8.204E+15	0.4	1.944E+17	0.3	9.928E+15	0.5
B-04-UB-4-CS-1	30-AUG-84	20:23:17	1.132E+18	0.3	2.334E+17	0.3	1.135E+18	0.6
B-04-UB-1-CS-1	30-AUG-84	19:36:38	1.129E+18	0.3	2.316E+17	0.3	1.147E+18	0.8

ZIRCONIUM/CERIUM SAMPLE RESULT TABLE FOR ONE ROD ON 12 NOON 01-JAN-84

SAMPLE I.D.	ANALYSIS DATE	ANALYSIS TIME	CE-144 ATM/G	CE-144 RSD	ZR-95 ATM/G	ZR-95 RSD
B-02-UB-1-ZR-1	24-AUG-84	20:15:42	5.555E+15	0.2	4.021E+13	2.4
B-02-UB-4-ZR-1	24-AUG-84	20:42:10	5.890E+15	0.2	4.705E+13	2.8
B-02-UB-1-ZR-2	29-AUG-84	18:29:42	5.751E+15	0.1	4.729E+13	1.8
B-02-UB-4-ZR-2	29-AUG-84	19:18:43	5.780E+15	0.1	4.731E+13	1.7
B-03-UB-1-ZR-2	29-AUG-84	20:52:09	9.588E+15	0.1	7.222E+13	2.9
B-03-UB-4-ZR-1	29-AUG-84	22:20:44	9.713E+15	0.1	7.356E+13	2.7
B-01-UB-1-ZR-1	29-AUG-84	23:47:28	4.789E+13	0.3	5.162E+11	2.9
B-01-UB-4-ZR-1	30-AUG-84	01:12:23	4.977E+13	0.3	5.616E+11	2.7
B-06-UB-1-ZR-1	09-SEP-84	00:18:14	3.398E+15	0.1	8.203E+12	5.1
B-06-UB-4-ZR-1	09-SEP-84	15:25:55	2.550E+15	0.1	6.312E+12	5.4
B-04-UB-1-ZR-1	10-SEP-84	01:01:12	1.121E+16	0.1	8.138E+13	2.2
B-04-UB-4-ZR-1	10-SEP-84	05:27:27	1.094E+16	0.1	7.894E+13	2.2
B-07-UB-1-ZR-1	10-SEP-84	11:24:16	5.798E+13	0.1	1.071E+11	4.6
B-07-UB-4-ZR-1	10-SEP-84	18:23:57	5.771E+13	0.1	9.650E+10	5.3
B-05-UB-1-ZR-1	07-SEP-84	22:59:43	8.165E+15	0.1	4.239E+13	3.0
B-05-UB-4-ZR-1	08-SEP-84	12:04:12	7.534E+15	0.1	3.993E+13	3.0

Fig. 26. Examples of Summary Data Files Containing Information from Gamma-Counting of Dissolver-Solution Aliquots:

- File of Results for CS Aliquots from Rod B and
- File of Results for ZR Aliquots from Rod B.

of the operations involved in calculating and reporting the results that were obtained.

In the mass spectrometric determination of gas composition, a low pressure (usually around a few hundred microns) of the sample gas is introduced into the mass spectrometer source, and molecules of the gas components are ionized, in our case by a beam of electrons. The positive ions formed in this way are separated according to their mass-to-charge (m/e) ratio, and a mass spectrum consisting of a plot of ion intensity vs. m/e value is recorded. Components in the sample are identified by their characteristic mass spectral patterns, i.e., the relative numbers of ions at specific m/e values. Such patterns exist for both simple and complex molecules as a result of multiple ionization and fragmentation of polyatomic species. The number of ions at a given m/e value

(i.e., the peak height at a particular mass number) is proportional to the partial pressure of the species in the source that contribute to the peak. Thus, the heights of peaks in the spectrum may be related to the partial pressure of specific components of the sample in the mass spectrometer source.

In our EOL operations, heights of relevant peaks from the mass spectrum were measured from a strip-chart recording and were manually tabulated and interpreted to identify components of the sample gas. The partial pressure of each component, P_x , was then calculated from the peak height, I_x , of the largest peak (the "base peak") in the pattern arising from the given component as follows:

$$P_x = S_x I_x \quad (\text{IV-13})$$

The sensitivity coefficient, S_x , was determined from prior analysis of pure gases and gas mixtures containing components of potential interest. When ion fragments or multiply charged ions from a higher-mass component coincided with the base peak of a lower-mass constituent, or when two or more components had the same base peak, contributions to I_x from the various components were resolved according to intensities of minor peaks in the pattern from each suspected contributor. When all peaks apparent in a spectrum were assigned and partial pressures of the individual components were calculated, validity of these results was tested by comparing the sum of the calculated partial pressures with the source pressure in the mass spectrometer. Agreement between these values provided confidence that no important peaks in the spectrum had been overlooked or misinterpreted during the spectrum analysis.

The volume percent of each identified component in the gas sample was calculated from its partial pressure and the sum of partial pressures of all components by means of the relationship

$$C_i \text{ (vol \%)} = (P_i / \sum P_x) 100\% \quad (\text{IV-14})$$

where C_i is the volume-percent concentration of the specified component, P_i is its partial pressure, and the summation is taken over all identified components.

Because the sensitivity coefficients for all isotopes of a heavy element such as Xe or Kr are identical, the handling of mass spectrometric data for determination of the isotopic composition of the fission gases is simpler than that required for determining the composition of a mixture of different gases. For the isotopic analysis of Kr and Xe in the POB gas samples, the mole percent of each isotope was calculated from the ratio of the peak height observed for the given isotope to the sum of corresponding peak heights for all isotopes of the element. A relationship analogous to that in Eq. IV-14 was used for this calculation.

Compositions of the POB gas samples were normally determined with the CEC 21-620 cycloidal mass analyzer (Section III.B.4). However, isotopic measurements on Xe and Kr in the plenum-gas samples were performed with the Nuclide RMS-7 instrument, which shows better resolution of the isotopic species

and somewhat greater sensitivity for these elements. While the plenum-gas samples were in the Nuclide RMS-7 source for the isotopic measurements, their entire spectrum was recorded and analyzed to corroborate the CEC 21-620 measurements. Agreement between the compositions measured on the two separate units provided confidence that errors introduced by calibration drift or other sources were negligible.

Results of the mass spectrometric analyses of the individual gas samples were entered into the project data system by means of a data-entry program that was written specifically for this purpose. The program prompted the analyst to enter results for specific components anticipated for each sample and wrote the data to one of three types of data file, depending on the sample identification and the data that were entered. The files contained results of either plenum-gas composition measurements, or isotopic measurements on Xe and Kr in the plenum gases, or composition data for the off-gas collected during dissolution of segments from a given fuel rod. The content of each of these files is illustrated in Figs. 27-29.

These files were subsequently accessed by project report-generator programs, which reformatted the data for samples related to a specified rod and used the data to compute results pertaining to fission-gas release during rod processing. The fission-gas results are discussed in Section V.G.

SAMPLE I.D.	XE	NR	H2	HE	VOLUME PERCENT		CO2	AR	CO2	NOX	OTHER	ANALYST INITIALS	ENTRY DATE
					H2O	N2							
A-00-CS-1	7.400E+00	1.600E+00	0.0	B4.5	0.0	4.9	1.4	0.1	0.0	0.0	0.0	MI	12-DEC-83
B-00-CS-2	2.00E-01	6.00E-01	0.0	B4.5	0.0	9.1	2.5	0.1	0.0	0.0	0.0	TCE/PH	22-AUG-84
C-00-CS-1	1.20E-01	3.00E-02	0.0	0.8	0.0	0.3	0.0	0.0	0.0	0.0	0.0	TCE/DC	19-SEP-84
(0R-)	7.500E-01	2.100E-01	0.0	55.2	0.1	33.7	9.6	0.0	0.1	0.0	0.0	TCE/DC	19-SEP-84
C-00-CS-2	2.00E-02	1.00E-02	0.0	0.5	0.0	0.3	9.2	0.0	0.0	0.0	0.0	TCE/DC	19-SEP-84
(0R-)	8.300E-01	2.200E-01	0.0	57.4	0.1	31.9	9.2	0.4	0.0	0.0	0.0	TCE/DC	19-SEP-84
D-00-CS-1	2.00E-02	1.00E-02	0.0	0.6	0.0	0.3	0.1	0.0	0.0	0.0	0.0	TCE/DC	29-OCT-84
(0R-)	2.00E-01	5.200E-02	0.0	91.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	TCE/DC	29-OCT-84
D-00-CS-2	2.00E-01	5.500E-02	0.0	96.4	0.0	2.6	0.7	0.0	0.0	0.0	0.0	TCE/DC	13-NOV-84
(0R-)	2.00E-02	5.00E-03	0.0	1.0	0.0	0.2	0.1	0.0	0.0	0.0	0.0	TCE/DC	10-DEC-84
E-00-CS-1	4.300E-01	1.100E-01	0.0	93.8	0.0	4.3	1.2	0.1	0.0	0.0	0.0	TCE/DC	10-DEC-84
(0R-)	2.00E-02	1.00E-02	0.0	96.2	0.0	3.1	1.0	0.0	0.0	0.0	0.0	9g/dg	10-DEC-84
F-00-CS-1	3.600E-01	2.00E-02	0.0	98.2	0.0	0.1	0.1	0.0	0.0	0.0	0.0	9g/dg	10-DEC-84
(0R-)	3.500E-01	9.100E-02	0.0	96.4	0.1	3.1	0.9	0.0	0.0	0.0	0.0	9g/dg	10-DEC-84
F-00-CS-2	2.00E-02	5.00E-03	0.0	91.8	0.0	3.9	0.9	0.0	0.0	0.0	0.0	TCE/DC	4-JAN-85
(0R-)	2.00E-02	6.300E-01	0.0	91.8	0.0	0.1	1.2	0.0	0.0	0.0	0.0	TCE/DC	4-JAN-85
G-00-CS-1	1.20E-01	6.200E-02	0.0	93.0	0.0	3.0	1.0	0.0	0.0	0.0	0.0	TCE/DC	4-JAN-85
(0R-)	2.320E+00	2.00E-02	0.0	92.1	0.0	5.4	1.8	0.0	0.0	0.0	0.0	TCE/DC	29-JAN-85
G-00-CS-2	5.00E-01	1.500E-02	0.0	92.1	0.0	0.9	0.1	0.0	0.0	0.0	0.0	TCE/DC	29-JAN-85
(0R-)	9.00E-03	5.00E-03	0.0	92.2	0.0	5.9	0.1	0.0	0.0	0.0	0.0	TCE/DC	28-FEB-85
H-00-CS-1	1.00E-02	5.900E-01	0.0	94.0	0.0	2.3	0.1	0.0	0.0	0.0	0.0	TCE/DC	29-JAN-85
(0R-)	2.200E+00	2.00E-02	0.0	92.9	0.0	4.1	1.2	0.1	0.0	0.0	0.0	TCE/DC	28-FEB-85
H-00-CS-2	1.00E-01	1.00E-02	0.0	92.9	0.0	0.2	0.1	0.0	0.0	0.0	0.0	TCE/DC	28-FEB-85
(0R-)	1.00E-01	2.700E-01	0.0	92.4	0.0	4.8	0.2	0.0	0.0	0.0	0.0	TCE/DC	28-FEB-85
J-00-CS-1	5.00E-02	2.00E-02	0.0	92.9	0.0	0.4	0.0	0.0	0.0	0.0	0.0	TCE/DC	19-MAR-85
(0R-)	1.100E+00	2.700E-01	0.0	92.9	0.0	0.1	1.2	0.1	0.0	0.0	0.0	TCE/DC	19-MAR-85
K-00-CS-1	1.00E-02	3.00E-02	0.0	74.0	0.0	19.2	5.2	0.2	0.0	0.0	0.0	TCE/DC	22-APR-85
(0R-)	1.00E-01	2.400E-01	0.0	68.8	0.0	0.4	6.2	0.0	0.0	0.0	0.0	TCE/DC	22-APR-85
K-00-CS-2	8.200E-01	3.00E-02	0.0	42.2	0.0	4.6	12.0	0.5	0.0	0.0	0.0	TCE/DC	22-MAY-85
(0R-)	4.400E-01	1.300E-01	0.0	42.2	0.0	9.4	9.4	0.4	0.0	0.0	0.0	TCE/DC	25-JUN-85
L-00-CS-1	1.40E-01	7.800E-02	0.0	82.1	0.0	0.3	3.0	0.1	0.0	0.0	0.0	TCE/DC	25-JUN-85
(0R-)	3.030E+00	1.50E-01	0.0	78.9	0.0	0.2	0.1	0.0	0.0	0.0	0.0	TCE/DC	18-JUL-85
M-00-CS-1	1.570E+00	4.200E-01	0.0	74.9	0.0	1.4	0.1	0.0	0.0	0.0	0.0	TCE/DC	18-JUL-85
(0R-)	1.360E-02	3.600E-01	0.0	65.8	0.0	25.3	6.8	0.3	0.0	0.0	0.0	TCE/DC	18-JUL-85
M-00-CS-2	7.00E-02	4.00E-02	0.0	74.0	0.0	0.4	5.2	0.2	0.0	0.0	0.0	TCE/DC	30-JUL-85
(0R-)	1.340E+00	3.400E-01	0.0	0.5	0.0	18.8	0.2	0.0	0.0	0.0	0.0	TCE/DC	30-JUL-85
N-00-CS-1	8.00E-02	3.00E-02	0.0	74.8	0.0	0.3	0.1	0.0	0.0	0.0	0.0	TCE/DC	30-JUL-85
(0R-)	1.410E+00	3.500E-01	0.0	91.8	0.1	12.8	5.2	0.1	0.0	0.0	0.0	TCE/DC	21-AUG-85
O-00-CS-2	1.100E-01	4.500E-01	0.0	76.8	0.1	17.6	5.2	0.2	0.0	0.0	0.0	TCE/DC	21-AUG-85
(0R-)	1.10E-01	4.00E-02	0.0	88.1	0.0	0.3	6.7	0.0	0.0	0.0	0.0	TCE/DC	21-AUG-85
P-00-CS-1	1.430E+00	3.600E-01	0.0	90.7	0.0	27.7	8.2	0.0	0.0	0.0	0.0	TCE/DC	4-SEP-85
(0R-)	3.210E+00	8.400E-02	0.0	59.7	0.0	0.4	0.1	0.0	0.0	0.0	0.0	TCE/DC	4-SEP-85
Q-00-CS-1	2.00E-01	9.100E-01	0.0	63.0	0.1	25.8	6.4	0.3	0.0	0.0	0.0	TCE/DC	4-SEP-85
(0R-)	2.00E-01	5.00E-02	0.0	87.3	0.0	9.8	2.7	0.1	0.0	0.0	0.0	TCE/DC	30-SEP-85
R-00-CS-1	1.800E-02	1.00E-03	0.0	0.9	0.0	0.2	0.1	0.0	0.0	0.0	0.0	TCE/DC	30-SEP-85
(0R-)	1.500E-02	1.00E-03	0.0	78.7	0.0	16.5	4.5	0.0	0.0	0.0	0.0	TCE/DC	30-SEP-85
R-00-CS-2	3.00E-03	4.00E-03	0.0	0.8	0.0	0.3	0.1	0.0	0.0	0.0	0.0	TCE/DC	30-SEP-85

Fig. 27. Data File Containing Results of Mass Spectrometric Measurements to Determine Plenum-Gas Compositions

SAMPLE I.D.	XE ISOTOPES				Kr ISOTOPES				ANALYST INITIALS		ENTRY DATE
	128	130	131	132	82	83	84	85	86		
A-00-05-1	0.1	0.2	11.3	20.1	0.2	16.2	30.5	3.8	49.3	PH	12-DEC-83
A-00-05-2	0.1	0.2	10.2	0.2	0.2	0.3	0.2	0.1	0.3	PH	12-DEC-83
B-00-05-1	0.0	0.2	11.2	20.6	0.2	16.3	30.3	3.8	49.4	TGE	20-AUG-84
B-00-05-2	0.0	0.1	0.1	0.2	0.2	0.2	0.2	0.2	0.2	TGE	19-SEP-84
C-00-05-1	0.1	0.0	12.4	22.6	0.2	15.4	30.1	6.1	48.2	TGE/DG	19-SEP-84
C-00-05-2	0.1	0.1	12.4	22.6	0.1	15.4	29.8	6.0	48.6	TGE/DG	19-SEP-84
D-00-05-1	0.0	0.0	13.9	22.3	0.0	16.4	0.2	6.0	0.2	TGE/DG	29-OCT-84
D-00-05-2	0.0	0.0	0.2	0.2	0.0	0.2	0.4	0.2	0.4	TGE/DG	29-OCT-84
E-00-05-1	0.0	0.0	13.9	22.2	0.0	16.4	28.9	6.1	48.5	TGE/DG	13-NOV-84
E-00-05-2	0.0	0.0	0.2	0.2	0.0	16.4	29.7	6.1	48.5	TGE/DG	10-DEC-84
F-00-05-1	0.1	0.1	12.1	21.9	0.0	15.9	29.3	6.0	48.8	98/46	10-DEC-84
F-00-05-2	0.0	0.0	0.2	0.3	0.0	16.0	0.3	0.2	0.3	98/46	4-JAN-85
G-00-05-1	0.0	0.0	12.9	22.1	0.0	15.5	29.6	5.9	48.7	TGE/DG	4-JAN-85
G-00-05-2	0.0	0.1	0.2	0.3	0.0	15.5	0.2	0.1	0.3	TGE/DG	29-JAN-85
H-00-05-1	0.0	0.0	11.7	22.4	0.0	15.2	29.2	5.8	49.2	TGE/DG	29-JAN-85
H-00-05-2	0.0	0.0	12.5	21.2	0.0	15.7	29.1	6.1	49.1	TGE/DG	29-JAN-85
I-00-05-1	0.0	0.0	12.8	21.2	0.0	15.5	0.3	6.0	49.2	TGE/DG	29-JAN-85
I-00-05-2	0.0	0.1	0.2	0.2	0.0	15.3	0.3	0.2	0.4	TGE/DG	28-FEB-85
J-00-05-1	0.1	0.0	11.4	22.7	0.0	15.2	30.3	5.7	49.4	TGE/DG	28-FEB-85
J-00-05-2	0.0	0.0	10.2	22.3	0.2	14.6	30.5	6.0	48.7	TGE/DG	19-MAR-85
K-00-05-1	0.1	0.0	10.6	22.3	0.1	14.8	30.7	5.9	48.4	TGE/DG	19-MAR-85
K-00-05-2	0.0	0.2	11.8	22.7	0.2	15.6	0.2	0.1	0.2	TGE/DG	22-APR-85
L-00-05-1	0.0	0.0	11.8	22.7	0.1	15.6	29.6	5.8	48.6	TGE/DG	22-APR-85
L-00-05-2	0.0	0.0	10.1	22.2	0.2	15.2	30.3	5.2	49.3	TGE/DG	22-MAY-85
M-00-05-1	0.0	0.1	11.9	22.7	0.2	15.3	30.2	5.9	49.0	TGE/DG	22-MAY-85
M-00-05-2	0.0	0.0	11.1	22.6	0.2	15.2	30.0	5.6	49.0	TGE/DG	25-JUN-85
N-00-05-1	0.0	0.1	10.7	23.6	0.2	14.8	30.8	5.7	48.5	TGE/DG	25-JUN-85
N-00-05-2	0.1	0.1	11.8	24.0	0.2	14.9	30.7	5.7	48.4	TGE/DG	18-JUL-85
O-00-05-1	0.0	0.1	11.7	23.9	0.2	15.6	30.0	5.7	48.4	TGE/DG	18-JUL-85
O-00-05-2	0.0	0.0	0.1	0.2	0.2	15.7	0.1	0.1	0.2	TGE/DG	30-JUL-85
P-00-05-1	0.0	0.0	10.9	22.9	0.2	15.1	30.4	5.7	48.6	TGE/DG	30-JUL-85
P-00-05-2	0.0	0.0	0.2	0.2	0.2	15.2	30.2	5.9	48.7	TGE/DG	30-JUL-85
Q-00-05-1	0.0	0.0	10.8	22.9	0.2	15.1	30.5	5.7	48.5	TGE/DG	21-AUG-85
Q-00-05-2	0.0	0.0	10.6	23.2	0.2	15.1	30.5	5.7	48.5	TGE/DG	21-AUG-85
R-00-05-1	0.0	0.0	10.8	23.2	0.2	15.0	30.5	5.7	48.6	TGE/DG	4-SEP-85
R-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	4-SEP-85
S-00-05-1	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
S-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
T-00-05-1	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
T-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
U-00-05-1	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
U-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
V-00-05-1	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
V-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
W-00-05-1	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
W-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
X-00-05-1	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
X-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
Y-00-05-1	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
Y-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
Z-00-05-1	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85
Z-00-05-2	0.0	0.0	10.8	23.2	0.2	15.1	30.5	5.7	48.6	TGE/DG	30-SEP-85

Fig. 28. Data File Containing Results of Mass Spectrometric Measurements to Determine Xe and Kr Isotopy in Plenum-Gas Samples

SAMPLE I.D.	VOLUME PERCENT											ANALYST INITIALS	ENTRY DATE
	XE	KR	H2	HE	H2O	N2	O2	AR	CO2	NOX	OTHER		
B-02-GS-1	0.000E-02	2.000E-02	0.4	0.0	0.1	79.2	15.9	0.9	0.0	3.4	0.0	TE/DG	27-AUG-84
(+OR-)	0.000E-03	2.000E-03	0.1	0.0	0.1	0.4	0.2	0.1	0.0	0.3	0.0		
B-02-GS-2	7.200E-02	2.000E-02	0.4	0.0	0.1	77.4	17.2	0.9	0.0	3.9	0.0	TE/DG	27-AUG-84
(+OR-)	7.000E-03	2.000E-03	0.1	0.0	0.1	0.5	0.3	0.1	0.0	0.4	0.0		
B-01-GS-1	0.000E+00	0.000E+00	1.3	0.0	0.1	77.1	17.0	0.9	0.1	3.0	0.0	TE/DG	29-AUG-84
(+OR-)	1.000E-03	1.700E-03	0.1	0.0	0.0	0.5	0.3	0.1	0.0	0.4	0.0		
B-01-GS-2	0.000E+00	0.000E+00	1.2	0.0	0.1	77.2	17.1	0.9	0.1	3.4	0.0	TE/DG	29-AUG-84
(+OR-)	1.500E-03	1.500E-03	0.1	0.0	0.0	0.6	0.3	0.1	0.0	0.4	0.0		
B-03-GS-1	1.500E-01	3.500E-02	0.4	0.0	0.1	79.5	15.4	0.9	0.1	3.5	0.0	TE/DG	29-AUG-84
(+OR-)	1.000E-02	5.000E-03	0.1	0.0	0.0	0.7	0.3	0.1	0.0	0.4	0.0		
B-03-GS-2	1.400E-01	3.800E-02	0.4	0.0	0.1	79.0	15.7	0.9	0.1	3.0	0.0	TE/DG	29-AUG-84
(+OR-)	1.000E-02	5.000E-03	0.1	0.0	0.0	0.6	0.2	0.1	0.0	0.4	0.0		
B-04-GS-1	1.800E-01	4.500E-02	0.4	0.0	0.1	77.3	17.1	0.9	0.2	3.8	0.0	TE/DG	5-SEP-84
(+OR-)	1.000E-02	5.000E-03	0.1	0.0	0.0	0.8	0.3	0.1	0.1	0.4	0.0		
B-04-GS-2	1.800E-01	4.700E-02	0.4	0.0	0.1	77.0	16.5	0.9	0.4	4.5	0.0	TE/DG	5-SEP-84
(+OR-)	1.000E-02	5.000E-03	0.1	0.0	0.0	0.8	0.4	0.1	0.1	0.4	0.0		
B-05-GS-1	6.400E-02	1.700E-02	0.4	0.0	0.1	79.0	17.1	0.9	0.1	2.4	0.0	TE/DG	5-SEP-84
(+OR-)	3.000E-03	3.000E-03	0.1	0.0	0.0	0.8	0.4	0.1	0.1	0.4	0.0		
B-05-GS-2	6.300E-02	1.700E-02	0.4	0.0	0.1	79.4	16.7	0.9	0.1	2.3	0.0	TE/DG	6-SEP-84
(+OR-)	2.000E-03	2.000E-03	0.1	0.0	0.0	0.8	0.4	0.1	0.1	0.4	0.0		
B-07-GS-1	2.000E-03	1.000E-03	0.2	0.0	0.1	77.4	17.1	0.9	0.1	4.2	0.0	TE/DG	6-SEP-84
(+OR-)	2.000E-03	1.000E-03	0.1	0.0	0.0	0.7	0.3	0.1	0.1	0.4	0.0		
B-07-GS-2	1.000E-03	1.000E-03	0.2	0.0	0.0	76.5	17.8	0.9	0.1	4.5	0.0	TE/DG	10-SEP-84
(+OR-)	1.000E-03	1.000E-03	0.1	0.0	0.0	0.7	0.3	0.1	0.1	0.4	0.0		
B-05-GS-1	1.540E-01	3.800E-02	0.4	0.0	0.1	77.2	17.1	0.9	0.1	4.0	0.0	TE/DG	10-SEP-84
(+OR-)	1.200E-02	3.000E-03	0.1	0.0	0.0	0.7	0.3	0.1	0.1	0.4	0.0		
B-05-GS-2	1.520E-01	3.700E-02	0.4	0.0	0.1	74.9	17.2	0.9	0.2	6.2	0.0	TE/DG	10-SEP-84
(+OR-)	9.000E-03	3.000E-03	0.1	0.0	0.0	0.7	0.4	0.1	0.1	0.6	0.0		

Fig. 29. Data File Containing Results of Mass Spectrometric Measurements to Determine Dissolver Off-gas Composition. (File of data from Rod B segments is shown.)

V. DISCUSSION OF EOL RESULTS AND EVALUATION OF PERFORMANCE

As stated earlier, the EOL campaign at ANL involved the processing of 17 fuel rods from the LWBR. In addition, there was a mid-campaign performance verification in which a single test rod was fabricated at ANL from nonirradiated components provided by BAPL and then was processed with the same equipment and procedures as were employed for the LWBR fuel rods.

Results of ANL's measurements and destructive analyses on individual rods and rod segments were forwarded to BAPL at the completion of work on each rod. These reports included data from measurements performed on the whole (i.e., unsheared) rod, data describing the shearing of the rod into segments, and data from the chemical and radiometric analyses performed on the dissolved segments and on the gases released during shearing and dissolution operations. Each report also contained noteworthy observations recorded during ANL's processing of the corresponding fuel rod and detailed descriptions of any off-normal occurrences that took place while the rod or its segments were being processed. For reasons of space, these reports (totaling approximately 300 pages) are not reproduced here. Rather, in the interest of supporting the validity of the reported data, we present an analysis of errors associated with certain key results and summarize observations that apply to the EOL campaign as a whole and that might be of interest to others doing similar work. A listing of these reports is given in Attachment 4.

In the error analysis, random and systematic errors associated with specific operations are evaluated from data obtained during the EOL campaign or from pre-EOL characterization and qualification tests performed with the EOL systems; EOL data, when available, were always given preference in these evaluations. Uncertainties in reported results are estimated by standard error-propagation techniques and are used to demonstrate that, with minor exceptions, the project error requirements (see Section II.B) were met.

Specific areas evaluated include measurements of the unsheared rods (i.e., length and weight), shearing of the rods into segments and characterization of each segment, segment dissolutions, and the chemical and radiometric analyses performed to determine uranium isotopic composition, uranium loading, and fission-product loading and distribution (^{137}Cs , ^{144}Ce , ^{95}Zr , and fission gases). In each case, error sources that contribute to the uncertainty in specific results of the ANL destructive assay are discussed; the magnitudes of errors from each source are evaluated from appropriate data; and the errors are combined to provide estimates of the limit to probable bias and standard deviation for the appropriate result. The statistical parameters estimated in this way are then compared with the limits to allowable error specified by the project error requirements. In those few cases where requirements were not met, reasons for the discrepancies and their significance relative to overall LWBR-POB program goals are discussed.

A. Evaluation of Measurements on Unsheared LWBR Fuel Rods

1. Uncertainties in Measuring Fuel Rod Weight

Measurements of the weight of each LWBR fuel rod were described in Section IV.D.3. Bias in each fuel rod weighing was minimized by calibrating the

rod-weighing balance with an NBS-traceable standard weight immediately before weighing each rod. In addition, this bias was demonstrated to be less than the resolution (0.1 g) of the balance readout by the weighing of a transfer standard (see Section IV.D.1.b.) that was carefully compared to NBS-traceable weights outside the FSSF hot cell. Bias in measuring the fuel rod weight can, therefore, be taken to be negligible relative to the readability of the balance and can, at worst, be confidently stated to be less than 0.1 g. Since the limit to allowable bias in the fuel rod weighing, per project requirements, is 0.1 g, this requirement is considered to have been met.

Random error in the weighing of the fuel rods is best estimated from replicate rod-weighing data. A ready source of such data, which provides true replication of the entire weighing process, was obtained from the weighings made of each rod before and after the rod plenum was punctured. These rod weights may be considered duplicates because the amount of gas removed from the plenum was quite small. Removal of the helium and fission gas from the rod void volume and replacement of that gas with air were calculated to result in a weight gain by the rod of only a few milligrams, an amount imperceptible to the balance used for weighing these rods. Rod weighing data for the 17 fuel rods processed during the EOL campaign and the single test rod used in the mid-campaign performance verification are shown in Table 11. The near-zero average difference, -0.038 g, between the weights determined before and after puncture is not statistically significant and attests that both weighings measured a common rod weight. Thus, the weight values for each rod may be considered duplicates, and variations in these values for each individual rod are random. Because the separate weighings of each rod are indeed replicates, the standard deviation of an individual rod weighing may be calculated as the pooled standard deviation of this set of 18 duplicate weighings; the resulting standard deviation is 0.132 g. This level of imprecision does not meet the project requirement, which calls for a standard deviation smaller than 0.10 g. However, because the weights measured before and after plenum puncture both measure the same rod weight, the expected value for this weight may be calculated as the average of the two weighings, with a variance equal to $1/2$ of that for an individual weighing, i.e., a standard deviation of 0.093 g. Thus, if the average of the two rod weighings is taken as the best estimate of the true rod weight, the requirement on precision of the rod weight determination is met by the ANL data. It might also be argued that the standard deviation obtained with single rod weighings is acceptable because it differs so little from the required standard deviation.

We note that the statistics calculated from the data in Table 11 are strongly affected by the relatively large difference between the before and after puncture weights of Rod E. Treatment of this difference as an outlier in arriving at the rod-weight error estimates might be justified; however, such an approach would not change the conclusions reached by considering the data in their entirety.

2. Uncertainties in Measuring the Length of LWBR Fuel Rods

Measurement of the fuel rod length using the feed/measurement system on the full-scale shear was described in Section IV.D.5. As stated earlier, the rod length was obtained as the sum of the temperature-corrected gauge-rod length and the carriage displacement associated with moving the carriage from the gauge-rod end to the fuel-rod end. This carriage displacement was evaluated from the temperature-corrected AST readings and the REL readings obtained first with the

Table 11. Weights of LWBR Fuel Rods Measured Before and After Puncture of the Fuel Rod Plenum Region

Rod Designation	Weight Before Puncture, g	Weight After Puncture, g	Weight Difference (Before - After), g
B	3555.29	3555.40	-0.11
C	3555.93	3555.93	0.00
D	3555.05	3555.27	-0.22
E	3558.44	3558.94	-0.50
F	3568.42	3568.50	-0.08
G	3556.57	3556.38	+0.19
H	8560.25	8560.18	+0.07
I	4210.10	4209.92	+0.18
J	4194.28	4194.11	+0.17
K	4180.88	4181.12	-0.24
L	4219.58	4219.55	+0.03
M	1116.03	1115.95	+0.08
N	1114.56	1114.71	-0.15
O	1118.87	1118.95	-0.08
P	1115.71	1115.56	+0.15
Q	1114.02	1114.30	-0.28
R	8493.28	8493.21	+0.07
W ^a	2562.42	2562.38	+0.04
Average Difference:			-0.038 g
Pooled Standard Deviation:			0.132 g

^aRod W was a test rod used in a mid-campaign performance verification and did not come from the LWBR.

gauge rod and then with the fuel rod in position against the forward stop on the FSS feed/measurement system (cf. Eq. IV-6).

With this measurement scheme, errors propagate from two main sources: (1) errors in determining the length of the gauge rod, including errors associated with corrections for thermal expansion, and (2) errors associated with the displacement measured with the shear feed system. Error in the assigned value of the gauge rod length introduces a bias into the measured fuel rod length, while uncertainties in the thermal expansion corrections on the gauge rod length introduce random errors related to the random errors associated with the temperature measurements used to determine the correction. (The RTD sensors used in measuring rod temperatures in the FSSF were individually calibrated against thermocouples calibrated by the NBS and are, therefore, considered to be free of bias.) The length of the gauge rod used in the EOL rod-length measurements for all temperatures of interest is given by

$$L_{GRT} = (34.99643 \pm 0.00010 \text{ in.}) [1 + \alpha_{GR} (T_{GRA} - 20.0^\circ\text{C})] \quad (\text{V-1})$$

In this expression, L_{GRT} is the true gauge-rod length at temperature T_{GRA} , 34.99643 ± 0.00010 in. is the assigned gauge-rod length at 20°C as determined by ANL's Quality Assurance Division (QAD) using gauge blocks calibrated by the NBS, and α_{GR} is the coefficient of linear thermal expansion for the gauge rod material (carbon steel). The value of α_{GR} is 1.19×10^{-5} in./in.- $^\circ\text{C}$. If the temperature measurements are (conservatively) estimated to have a standard deviation, σ_T , of 0.1°C , then the standard deviation of the corrected gauge rod length may be estimated as 0.000042 in. by taking the product of σ_T and α_{GR} times 35 in. (the nominal gauge-rod length). A limit to the bias contribution from the gauge rod length to the determination of fuel rod length is given by the uncertainty, 0.0001 in., in the assigned value for gauge rod length.

Errors associated with the measurement of a displacement on the FSS feed system were characterized during pre-EOL development work with the FSS. Because of the precision required for the FSS measuring systems, an exceptionally reliable method for evaluating these errors had to be employed. A laser-based displacement monitoring system was used in the characterization tests. Details of the test results are given in Attachment 3. From the laser calibration of the systems, we determined that the displacement measurements would contribute a potential bias to the rod length values of -0.0003 in. and a standard deviation of 0.00126 in. Errors associated with the thermal expansion corrections applied to the AST readout are small relative to these other errors. For example, if the standard deviation for measuring the FSS machine-base temperature is 0.1°C and the linear expansion coefficient of the machine base is 2.5×10^{-5} in./in.- $^\circ\text{C}$, the random error introduced from this source into a measurement of a 120-in. rod length is only 0.0003 in. Adding these standard deviations in quadrature gives a total standard deviation for the displacement determination of 0.00130 in.

The potential bias that might exist in the rod length values reported for the EOL rods was estimated from the sum of the absolute value of the bias contribution (0.0001 in.) from the gauge rod length and that from the displacement measurement (0.0003 in.). Thus, a limit to potential bias in the rod length is estimated at 0.0004 in., which compares favorably with the limit to allowable bias, 0.001 in., prescribed by project error requirements.

Imprecision of the rod length measurements may be similarly estimated by summing the variances of these two parameters. Thus,

$$\begin{aligned} \sigma_{\text{Rod Length}}^2 &= \sigma_{GR}^2 + \sigma_{FSS}^2 \\ &= (0.000042 \text{ in.})^2 + (0.00130 \text{ in.})^2 \\ &= (0.00130 \text{ in.})^2 \end{aligned} \quad (\text{V-2})$$

where $\sigma_{\text{Rod Length}}^2$ is the variance of the rod length value, σ_{GR}^2 is the variance of the "true" gauge rod length, and σ_{FSS}^2 is the variance associated with a displacement measurement on the FSS. The standard deviation of the rod length measurement estimated in this way, 0.00130 in., is well within the allowable

random error, 0.005 in., permitted by project requirements. Hence, project requirements on the measurement of fuel rod length were met.

The above approach to estimating the uncertainties associated with the rod length determination implicitly assumes that the characteristics of the FSS feed/measurement systems did not change during the course of the EOL campaign. We are confident that such changes did not occur, based on observations made during measurements involving the gauge rod over the course of the campaign and long experience with the FSS prior to the EOL work. Nevertheless, it seemed prudent to acquire test data, following completion of the campaign, that would demonstrate the validity of this assumption. The test that was performed consisted of measuring the length of a stack of gauge rods, which were individually measured by ANL's QAD. (These gauge rods were used in the original pre-EOL laser-calibration tests performed on the FSS as described in Attachment 3.) The length of the stack was varied by combining several gauge rods, laid end to end, on the shear bed, and the length of the stack was measured in a manner analogous to that used for measuring the fuel rod lengths. The stack lengths measured with the FSS are compared in Table 12 with the corresponding lengths calculated from QAD calibration data for each gauge rod. The average difference between the measured and calculated length of each gauge rod stack was found to be 0.00013 in., which is consistent with the limit (0.0004 in.) to probable bias in the measurements determined from the laser-calibration data. The standard deviation of this difference (0.00034 in.) is somewhat smaller than that predicted from the laser-calibration data (0.00130 in.) and possibly reflects the fact that only a limited number of positions along the feed system were involved in the post-EOL demonstration. This result indicates that, at worst, the standard deviation estimated from the pre-EOL characterization might be somewhat conservative. It may be concluded with confidence, therefore, that the error requirements on the rod length measurements were met.

B. Evaluation of Performance during Shearing of LWBR Fuel Rods

Operations involved in the shearing of fuel rods during the EOL campaign were described in Section IV.D.6. Key parameters related to these operations included the locations of segment boundaries on each rod relative to the locations specified by BAPL, the lengths of the individual segments sheared from each rod, and fuel recovery during shearing. Discussed in the following subsections is the extent to which ANL met the error requirements for each parameter.

1. Evaluation of Performance in Locating Segment Boundaries

a. Factors Influencing the Location of Segment Boundaries

The equipment and procedures employed in the ANL shearing operations were designed to ensure precise positioning of the fuel rod for cutting at the locations specified by BAPL for segment boundaries. The actual location of the as-cut boundary was determined after each segment boundary cut was made. For each designated segment boundary, the fuel rod was positioned on the shear bed such that the cutting plane of the shear blade (the shear plane) was aligned with the desired location on the rod; the shear blade was lowered and the cut was made; and then, in effect, the length of rod that remained between the REL and shear plane was measured to determine the as-cut location of the

Table 12. Results of Post-EOL Test to Check Shear Feed System Performance

Trial No.	Gauge Rods in Stack ^a	Stack Temperature, °C	Calculated Stack Length, ^b in.	Measured Stack Length, in.	Difference (Measured - Calculated), in.
1	1	25.1	30.001695	30.004335	+0.002640 ^c
	1,2	25.1	48.003306	48.003246	-0.000060
	1,2,3	24.8	66.004652	66.005165	+0.000513
	1,2,3,4	25.3	85.006637	85.006424	-0.000213
	1,2,3,4,5	25.3	115.008510	115.009000	+0.000490
2	1	25.1	30.001695	30.004379	+0.002684 ^c
	1,2	25.2	48.003363	48.003334	-0.000029
	1,2,3	25.0	66.004809	66.005211	+0.000402
	1,2,3,4	25.3	85.006637	85.006271	-0.000366
	1,2,3,4,5	25.3	115.008510	115.008970	+0.000460
3	1	25.2	30.001730	30.004353	+0.002623 ^c
	1,2	25.2	48.003363	48.003277	-0.000086
	1,2,3	25.1	66.004888	66.005333	+0.000445
	1,2,3,4	25.3	85.006637	85.006302	-0.000335
	1,2,3,4,5	25.2	115.008380	115.008730	+0.000350
Average Difference:					+0.0001309 in.
Standard Deviation(s):					0.0003431 in.

^aGauge rods were laid end to end against a 30.000-in. gauge rod positioned at the forward stop on the shear feed system. Rod identifications in Note b.

^bCalculated by summing lengths of individual gauge rods corrected for thermal expansion using the formula $L = L_0 [1 + \alpha_{GR} (T_{GR} - 20^\circ\text{C})]$ where $\alpha_{GR} = 1.19 \times 10^{-5}$ in./in.-°C, T_{GR} is the measured stack temperature, and L_0 for each rod was as follows:

- 1: 29.999874 in.
- 2: 18.000519 in.
- 3: 18.000489 in.
- 4: 19.000394 in.
- 5: 29.999984 in.

^cThe end of the first 30.000-in. gauge rod falls in an area on the ball screw where a relatively large error in the length equivalent of the AST readout exists due to a dislocation in the lead of the ball-screw threads (see Attachment 3 for details). This area of the screw was avoided in EOL operation of the FSS, and the data for this position are not included in the analysis of the present test results.

boundary relative to the rod reference end (i.e. the end in contact with the REL).

Error requirements on the location of segment boundaries in shearing the LWBR fuel rods were stipulated not in terms of how accurately the actual locations of boundaries were to be measured at ANL, but rather in terms of how closely the boundaries fell relative to the locations specified on each rod's shearing diagram. Given the shearing procedure outlined above, coincidence of the desired and as-cut boundary locations for segments from a particular rod was found to depend on three major factors: (1) performance of the rod-positioning system in aligning the desired cut location with the shear plane; (2) performance of the rod-clamping system in maintaining the rod's position while the shear blade is being lowered; and (3) shearing behavior of the rod materials, especially with regard to any displacement occurring between the effective plane of separation of the material and the (nominal) plane defined by the shear-blade cutting edge as it traverses the rod's cross section. Each of these factors was extensively characterized during pre-EOL development of the FSS operations in which the test rods were composed of nonirradiated Zircaloy-cladding parts and high-density thorium pellets. Because of the importance of these factors in interpreting ANL's results from shearing the EOL fuel rods, we discuss them individually below.

(1) Positioning the Rod for Cutting

The feed system on the FSS was designed to position the rod for cutting at a specified boundary location in a manner equivalent to measuring the length of the rod remaining between the REL and the shear plane. It follows, then, that uncertainty in the rod position is identical to the uncertainty with which a given rod's length can be measured. In Section V.A.2, uncertainties associated with rod-length measurement performed with the FSS were shown to include a potential bias of 0.0004 in. and a standard deviation of 0.00130 in. These estimates of potential error in positioning a rod for cutting demonstrate that performance of the rod-positioning system was totally adequate to meet the requirements established for locating segment boundaries, if other significant error sources were absent. (The requirement specified that each boundary cut through the rod cladding must lie within 0.010 in. of the desired location; separate, less-stringent requirements applied for corresponding boundary cuts through the fuel.)

(2) Holding the Rod in Position While Cutting

Movement of the fuel rod during cutting was restrained by the spring-loaded clamp jaws on the carriage of the FSS and by the pneumatically operated hold-down positioners on the shear feed system. In the pre-EOL tests with nonirradiated rods, these restraints were shown to be sufficient to completely inhibit movement of the rod, except under conditions where a cut was made through solid Zircaloy with less than about 3 in. of the rod remaining to be cut. Such short rod sections could not be tightly held during the shear stroke because the closest hold-down positioner on the FSS was 3.5 in. from the shear blade, and the rod end could not be engaged in the clamp jaws. (As described in Section IV.D.6, the physical length of the carriage precluded its being indexed far enough forward to permit shearing of a short rod section locked in the clamp

jaws; a push rod was installed behind the rod section to accommodate this situation.) For such cuts, substantial movement (on the order of a few hundredths of an inch) of the rod section often occurred during the shear stroke, resulting in a correspondingly large displacement between the actual cut location and the desired value. On the basis of these pre-EOL observations, larger-than-ordinary error in locating the final boundary cut (i.e. the boundary cut through solid Zircaloy in the stub-end section) of any rod was expected and considered to be a normal consequence of the operations.

In many cases behavior of the LWBR fuel rods during shearing was not like that for nonirradiated rods; only the seed rods exhibited the nearly ideal behavior that nonirradiated test rods showed during the pre-EOL development work with the FSS. Shearing of the power-flattening-blanket, standard-blanket, and reflector rods was characterized by slippage of the rod in the carriage clamp jaws and occasional movement (kickback) of the spring-loaded clamp jaws during the shear stroke. In most instances, these movements were compensated for in the computerized shearing procedure and did not significantly affect the results of the shearing operations. When, however, kickback occurred during a segment boundary cut, a substantial deviation between the desired and actual boundary locations resulted. Rod movement during the shear stroke at a segment boundary thus represented a significant source of error in shearing the LWBR fuel rods, and thus has a significant impact in meeting the project requirement on making segment boundary cuts at the specified locations.

(3) Displacement of the Effective Shear Plane from its Nominal Position (Shear Plane Error)

A displacement between the effective plane of separation of rod materials relative to the nominal shear plane defined by the shear blade's cutting edge has been designated the "shear-plane error." Shear-plane errors in the cladding result mainly from tearing of the Zircaloy or distortion of the face of the cladding tube during the shear stroke. These cladding effects are generally quite small because the cladding is cut relatively cleanly. On the other hand, shear-plane error for ceramic fuel material is relatively large because the ceramic material is pulverized upon impact by the cutting blade, with the pulverization (and cracking) extending for a distance into the fuel region away from the blade's cutting plane.

Evidence for significant shear-plane displacement when ceramic fuel is sheared was first obtained with high-speed filming techniques during pre-EOL studies of the shearing process. For these studies, the faceplate of the shear-blade housing was replaced with a transparent plastic plate, and movement of the shear blade through the nonirradiated test-rod section containing high-density thoria pellets was filmed. The films obtained in this way showed that the ceramic material shatters early in the shear stroke and is expelled as finely divided, pulverized material from the end of the cladding tube that protrudes into the shear housing; after being sheared, this cladding section is referred to as a "hull." On film, we observed the cladding hull break free of the rod face before the blade completely covered the open end of the remaining rod. Also observed was a small amount of ceramic material falling into the shear-blade housing from the uncut portion of the rod (see selected film frames in Fig. 30).

Fig. 30. Selected Frames from High-Speed Motion Pictures of the Shearing Process

Subsequent to these observations, tests were carried out, again with nonirradiated thoria pellets, to derive a quantitative characterization of the shear-plane displacement in the ceramic fuel material. Details of these tests and results therefrom are described in Attachment 5. Based on these results, we estimated that the shear plane in the fuel material was displaced an average of 0.017 in. from the plane of the blade; the standard deviation of this displacement is 0.0048 in. In light of these observations, BAPL specified separate requirements on the location of segment boundaries in the fuel and in the cladding of the EOL LWBR fuel rods and on the measurement of the lengths for the fuel and cladding segments. The shear-plane error in each fuel segment was taken into account by BAPL as they laid out the shearing requirements for each LWBR fuel rod (cf. Table 2).

b. Errors in Locating Segment Boundaries Relative to Specified Locations

Deviations between the segment boundary locations specified for each LWBR fuel rod and the locations that were actually achieved are shown in Table 13. Each deviation listed in this table was calculated as the difference between the as-cut location reported by ANL and the location requested by BAPL. Because the ANL measurement corresponds to the location of the sheared end of the rod's cladding tube, these deviations apply in the case of segment boundaries in the cladding as opposed to segment boundaries in the fuel. Because the as-cut location of each boundary was determined with the same accuracy and precision (i.e., bias less than 0.0004 in., standard deviation of 0.00130 in.) as for a single measurement of rod length, these listed deviations may be considered as closely reflecting the performance of the FSS in locating the specified boundaries.

As expected, the largest deviations generally occur for the last boundary cut on each rod, i.e., the cut that was made through the solid Zircaloy stub section. As discussed in Section V.B.1.a.(2), the observed behavior for these cuts is normal and acceptable. Of the 135 segment boundaries in Table 13 that were not located in stub sections, eight fail to meet the requirement for locating boundary cuts in the cladding, which calls for a deviation not larger than 0.010 in. Thus, more than 94% of the segment boundaries were positioned within the allowable distance of the specified locations. Seven of the eight discrepant boundary locations occurred for cuts associated with one of the two reflector rods,

Table 13. Deviations Between As-Cut Locations of Segment Boundaries and Specified Locations for LWBR Fuel Rods

Boundary Designation	Deviations for Indicated Rod, mils ^a																
	B	C	D	E	F	G	H	I	J	K	L	M	N	O	P	Q	R
1	0	0	-1	0	0	0	0	0	-1	0	0	0	0	0	0	0	0
2	1	0	0	0	0	1	1	0	-1	2	0	-1	-2	0	0	0	0
3	-1	-2	0	1	0	0	-11	-1	0	1	0	0	0	1	0	-1	-1
4	-1	1	1	1	0	-1	13	1	0	1	1	0	-1	0	0	0	3
5	-1	-1	-1		0	0	1	0	1	1	2	0	1	0	-1	-1	-1
6	2	0	-1		2	0	-10	0	-2	16	-1	4	0	-1	-1	0	0
7	1	-1	0	-1	-1	1	6	1	0	0	-1	-2	45	38	25	16	0
8	25	49	25	27	0	35	2	-1	3	3	0	-	-	-	-	-	1
9	-	-	-	-	42	-	10	47	20	40	38	-	-	-	-	-	15
10	-	-	-	-	-	-	6	-	-	-	-	-	-	-	-	-	1
11	-	-	-	-	-	-	34	-	-	-	-	-	-	-	-	-	22
12	-	-	-	-	-	-	1	-	-	-	-	-	-	-	-	-	1
13	-	-	-	-	-	-	14	-	-	-	-	-	-	-	-	-	-2
14	-	-	-	-	-	-	14	-	-	-	-	-	-	-	-	-	0
15	-	-	-	-	-	-	-4	-	-	-	-	-	-	-	-	-	1
16	-	-	-	-	-	-	13	-	-	-	-	-	-	-	-	-	27

^aDeviation was obtained by subtracting the specified location from as-cut location. One mil = 0.001 in.

rods H or R. Possibly owing to radiation hardening of the Zircaloy cladding on these larger-diameter rods (0.832 in. as compared to 0.572 in. for the next smallest rod type), kickback of the reflector rods during shearing was exceptionally frequent and severe and represented the primary reason for the large discrepancies that were encountered. Because the boundary location errors that occurred during the EOL campaign were few and resulted from the unpredictable shearing behavior of the irradiated fuel rods, BAPL indicated early in the campaign that performance of the FSS in locating the segment boundaries was acceptable on a best-effort basis. Moreover, because the actual locations of the segment boundaries were reliably known from the ANL measurements, the deviations between requested and as-cut locations could be accounted for in BAPL's final evaluation of the shearing data. Thus, although not on a par with what was expected on the basis of pre-EOL tests with nonirradiated rods, performance of the FSS in locating segment boundaries with the LWBR fuel rods was satisfactory overall when boundary locations in the cladding were considered.

As-cut locations for segment boundaries in the fuel material of each rod could not be determined from the EOL shearing data and, therefore, had to be inferred from the cladding boundary locations and the pre-EOL data characterizing the fuel's shear-plane error. This inference requires that one assume identical shearing behavior for the irradiated and nonirradiated ceramic materials. In estimating the uncertainty with which the boundary cuts in the fuel material may be determined, we had to combine uncertainties associated with locating the cut through the cladding and uncertainties associated with parameters describing the fuel's shear-plane error. Thus, for each segment boundary in the fuel, a bias term (equal to any difference that might exist between the true value of the average shear-plane error for the fuel and the value used in computing the cut location in the fuel) must be added to the estimated potential bias in the cut location determined for the cladding. The magnitude of such a bias term is difficult to estimate, since the proper value for the shear-plane displacement cannot be exactly determined. For the purpose of this analysis, we chose to assign a limit to its magnitude equal to the half-width of the 95% confidence interval about the average fuel shear-plane error as determined from the test data in Attachment 5. The average fuel shear-plane displacement was computed as the average of 19 separately measured displacements in the shear plane having a standard deviation, s , equal to 0.0048 in. and 13 degrees of freedom. Thus, the half-width, W , is given by the expression

$$W = (t_{0.05,13}) (s/\sqrt{n}) = (2.18)(0.0048 \text{ in.})/\sqrt{19} = 0.0024 \text{ in.} \quad (\text{V-3})$$

A limit to potential bias in the fuel-segment boundary location, as measured with the FSS and corrected for the fuel shear-plane displacement, is the arithmetic sum of the bias contributions from these two operations, i.e., 0.0004 in. plus 0.0024 in., or 0.0028 in.

The variance, $\sigma_{\text{BCL,Fuel}}^2$, of the calculated segment boundary locations in the fuel is likewise obtained by summing the variance of the FSS measurement and the variance of the fuel shear-plane error (designated σ_{FSS}^2 and $\sigma_{\text{SP,Fuel}}^2$, respectively). Thus,

$$\begin{aligned}
 \sigma_{\text{BCL,Fuel}}^2 &= \sigma_{\text{FSS}}^2 + \sigma_{\text{SP,Fuel}}^2 \\
 &= (0.00130 \text{ in.})^2 + (0.0048 \text{ in.})^2 \\
 &= (0.0050 \text{ in.})^2
 \end{aligned}
 \tag{V-4}$$

The standard deviation of the calculated fuel-segment boundary location is thus estimated to be 0.0050 in.

The potential bias (0.0028 in.) and the standard deviation (0.0050 in.) estimated in this way for the as-cut locations of segment boundaries in the fuel material compare quite favorably with the limits (0.005-in. bias, 0.015-in. standard deviation) to allowable error established in the project requirements. Thus, if as-cut locations of the segment boundaries are used in BAPL's evaluation of ANL's shearing data, this requirement will have been satisfied.

2. Evaluation of Performance in Determining Segment Lengths in Cladding and Fuel

Individual segment lengths reported by ANL for the LWBR fuel rods were computed as the difference between the locations of the as-cut boundaries that defined the two ends of the sheared segment. These reported segment lengths thus correspond to the cladding length of the segment. For all fuel-bearing segments in a given rod, except those involving the ends of the fuel stack, the reported length also corresponds to the fuel segment length, since the boundary location in the fuel may be assumed to be displaced by a constant amount from the location on the cladding as a result of the fuel shear-plane error. Thus, the fuel segment is displaced (by a nominal 0.017 in.) from the cladding segment but will have the same length.

For the two fuel-bearing segments that involved the fuel stack ends (always the first and last fuel-bearing segments for a given rod), only one end of the segment was defined by a shear cut through the fuel; the other end was taken to correspond to the fuel stack end, the location of which was established by BAPL apart from the work at ANL. Uncertainties in the fuel segment length for these cases depend on the manner in which BAPL evaluated the locations of the stack ends, and the uncertainties associated with their evaluation. Since ANL was not involved in making these calculations, we cannot estimate the errors that relate to them. Our discussion of the uncertainties associated with the segment length determinations will, therefore, be restricted to cladding segments and to fuel segments for which the fuel stack was sheared at both segment ends.

Bias errors in the measured boundary locations that were used to calculate the reported segment lengths were discussed in Section V.B.1.b. When the segment length is calculated as the difference between two boundary locations, these bias errors cancel in the calculation because they are virtually independent of the location of the segment boundary or the length of the segment under consideration. This cancellation occurs regardless of whether fuel segment length or cladding segment length is being considered. Relative to the error requirements on these segment lengths, then, probable bias in the results is inconsequential (limits to allowable bias are 0.001 in. for cladding segment length and 0.005 in. for fuel segment length). Thus, requirements on bias in the segment length determinations are easily met by the ANL data.

The variance of the calculated segment length may be similarly estimated by propagating the variances of the measured boundary locations used in the calculation. As shown in Section V.B.1.b, the magnitude of the boundary location variance is different for boundaries in the fuel and in the cladding, but it is the same for all measured boundaries in one or the other material. From standard error propagation formulas, the variance of a difference between two statistically independent variables is given by the sum of the variances of the individual variables. In the case being considered, where both measured variables have the same statistically independent variance, one may write

$$\sigma_{\text{DIFF}}^2 = \sigma_x^2 + \sigma_x^2 = 2\sigma_x^2 \quad (\text{V-5})$$

where σ_{DIFF}^2 is the variance of the difference, and σ_x^2 is the variance of the measured variable. For a cladding-segment length determination, the standard deviation of the boundary locations was estimated earlier as 0.00130 in. The corresponding standard deviation of a difference between two boundary locations (i.e., the standard deviation of the cladding segment length), from Eq. V-5, is 0.00184 in.

In the pre-EOL development work on characterizing the FSS performance with nonirradiated materials, a standard deviation of 0.0019 in. was observed for lengths of short cladding segments; this work was part of that described in Attachment 4. The close agreement between that experimental value and the value derived here from consideration of the laser calibration performed on the shear feed system supports the validity of the present treatment.

The standard deviation of the cladding segment length determinations (0.00184 in.) estimated for the ANL EOL data is smaller than the limit (0.005 in.) allowed by the project error limits. Thus, our analysis of errors based on our best estimates of the various contributions to the errors in the cladding-segment length determinations indicates that this requirement was met.

By assigning the appropriate value from Section V.B.1.b to the standard deviation of a fuel-segment boundary location and following an analogous treatment to that for the cladding segments, we estimated the standard deviation of the fuel-segment length determinations as 0.0071 in. This estimated value is considerably smaller than the limiting standard deviation (0.015 in.) allowed by the error requirements and indicates that this requirement was also satisfied.

3. Evaluation of Performance in Recovering Sheared Fuel

Fuel recovery during shearing of each individual LWBR fuel rod was evaluated by BAPL using information obtained during the shearing operations at ANL. This information included the weight of the unsheared rod and the weights of each segment collected when the rod was sheared. Specific error requirements were established regarding the allowable bias and standard deviation associated with the determination of each of these weights. Uncertainties in ANL's determination of the weight of the unsheared rod were discussed in Section V.A.1. Uncertainties on the segment weights are considered below, in Section V.B.3.a.

Because the effect of irradiation on the shearing behavior of fuel material (and hence on fuel loss) was recognized as being unpredictable, acceptance criteria for fuel recovery during shearing were specified in the project error requirements in terms of minimum acceptable performance levels based on studies involving nonirradiated materials. These specifications called for a recovery fraction of the fuel material from the whole rod not less than 99.75% and an accurate determination of the standard deviation of the individual segment fuel-recovery fraction, which was not to exceed 0.25%. Pre-EOL studies with nonirradiated test rods demonstrated that, in fact, fuel recovery during shearing of thorium fuel pellets consistently exceeded 99.9%. Determination of the standard deviation of the segment fuel-recovery fraction was carried out as part of the qualification of the FSSF¹⁴; the value obtained for this parameter was 0.0758%, well within the allowable range.

In effect, fuel recovery in shearing irradiated LWBR rods was on a best-effort basis, but it was hoped that the level of performance would be at least consistent with the 99.75% minimum criterion applied to the nonirradiated test rods. That this hope was justified is demonstrated in Section V.B.3.b, where fuel-recovery data obtained for fuel rods sheared during the EOL campaign are presented and discussed.

a. Uncertainty in the Determinations of Segment Weight

Weights of the individual rod segments collected during shearing of the LWBR fuel rods were calculated as the difference between the measured weight of a particular aluminum sample can when it contained the segment and when it was empty. Uncertainty in the calculated segment weight may, therefore, be estimated by propagating the errors associated with these full- and empty-can weighings. As stated in Table 2, the allowable bias for segment weights is 0.001 g if the weight is <286 g and 0.01 g otherwise. All of the empty aluminum cans (actually, each can and its cap were weighed together) were weighed on the same 300-g capacity balance (resolution, 0.001 g) in the FSSF. Full cans weighing less than 300 g (segment weight less than 286 g) were also weighed on this balance, whereas those weighing 300 g or more were weighed on a 2000-g capacity balance, which had a resolution of 0.01 g. Both of these balances were calibrated inside the hot cell against an NBS-traceable standard weight, immediately prior to their use for each rod, and the validity of this calibration was demonstrated by means of a transfer standard (see Section IV.D.1.b) that was carefully compared to NBS-traceable weights outside the cell. The computerized weighing procedure used in these comparisons required that the in-cell and out-of-cell weights measured for the transfer standard not differ by more than the balance resolution. On the basis of this comparison, bias errors due to miscalibration of the FSSF balances are presumed to be negligible relative to the readability of the balance and can, at worst, be confidently stated to be less than the balance resolution.

When the segment weight is calculated from full- and empty-can weighings performed on the same balance, residual bias errors cancel. Thus, for segments weighing less than 286 g, a second argument besides that based on the use of transfer standards indicates that bias in the segment weight may be considered inconsequential relative to the limit (0.001 g) allowed by project error requirements. For segments weighing 286 g or more, the bias associated with the

larger balance applies, and the segment weight value has a maximum probable bias of 0.01 g. Such a bias error is still sufficient to meet the requirement (0.01 g) for bias in the weight determination for segments heavier than 286 g. This analysis shows, therefore, that the requirements on bias in the segment weight determinations were met.

Random error in the segment weight determinations can likewise be estimated from random errors associated with the measured full- and empty-can weights. To evaluate these random errors, we chose to treat as duplicate measurements (1) the in-cell and out-of-cell weighings performed on each empty aluminum can and (2) the full-can weighings that were performed in the FSSF and DDS hot cell for each segment. The corresponding weighings in each of these locations were performed using essentially identical balances, which, for the data analysis, were presumed to exhibit the same levels of imprecision, i.e., to share a common standard deviation for weighings performed with them.

The in-cell and out-of-cell weighings of the empty cans provided 169 pairs of measurements showing an average difference between the in-cell and out-of-cell values of 0.0011 g and a standard deviation for a single measurement of 0.0013 g. The FSSF and DDS weighings of the full cans gave 52 pairs of measurements for full cans weighing less than 300 g; these data showed an average difference of -0.0022 g and a standard deviation for an individual measurement of 0.0034 g. There were 100 pairs of weighings for full cans heavier than 300 g, which showed an average difference of 0.0082 g and a standard deviation for an individual measurement of 0.0062 g.

The non-zero values that were obtained for the average differences from each of these data sets may indicate that small biases did exist between the different balances used for the measurements. In only one of the three cases, however, is the average difference greater than the balance resolution, i.e., the -0.0022 g difference between full-can weights measured in the FSSF and DDS hot cell. We attribute this difference to errors in the weighings associated with the DDS, possibly caused by slight deterioration of the standard weights at the DDS as a result of corrosive acid vapors in the DDS cell atmosphere. Weighing errors resulting from the use of these standards were noted during the EOL campaign, but they did not exceed tolerances established for the various DDS weighing operations because these were not as stringent as the tolerances required of the FSSF weighing systems. The magnitude of the observed difference between the FSSF and DDS full-can weights is thus consistent with expectation and is not taken as evidence that the bias requirement on the segment weight determination was not met.

The standard deviation of the segment weight determinations, $\sigma_{\text{Seg. Wt.}}$, was estimated from the standard deviations of the full- and empty-can weighings, $\sigma_{\text{Full Can}}$ and $\sigma_{\text{MT Can}}$, respectively, according to the relationship

$$\sigma_{\text{Seg. Wt.}}^2 = \sigma_{\text{Full Can}}^2 + \sigma_{\text{MT Can}}^2 \quad (\text{V-6})$$

For segments weighing less than 286 g, $\sigma_{\text{Full Can}}$ was taken as 0.0034 g and $\sigma_{\text{MT Can}}$ as 0.0013 g, based on the data presented above. The value obtained for $\sigma_{\text{Seg. Wt.}}$ from Eq. V-6 in this case is 0.0036 g, which is well

within the limit to allowable random error (0.005 g) for the segment weight determination. Thus, the requirement on segment weight was met for these segments.

For segments weighing more than 286 g, $\sigma_{\text{Full Can}}$ was taken as 0.0062 g and $\sigma_{\text{MT Can}}$ kept the same value as for the lighter segments. In this case, $\sigma_{\text{Seg. Wt.}}$ is calculated to be 0.0063 g. This estimated value for $\sigma_{\text{Seg. Wt.}}$ is slightly larger than the limit (0.005 g) allowed by project error requirements. We knew at the outset of the EOL campaign that this particular requirement, which calls for a standard deviation equal to only 1/2 the resolution of the balance used for the weighings, would be difficult to meet. Because pre-EOL characterization testing of the FSSF weighing systems indicated that meeting the requirement was probable, however, ANL did not request its re-evaluation by BAPL. In spite of not meeting this requirement, we believe that this observed standard deviation for the determination of segment weights greater than 286 g is the best that could be achieved with equipment that was commercially available when the ANL facilities were set up. Moreover, this standard deviation corresponds to a relative standard deviation of only 0.0022% for a segment weighing the minimum 286 g (worst case). As such, it is hard to imagine that this error would affect the ability of the ANL data to meet the DOE-NR parametric requirements (see Table 1) to which errors in the segment weight contribute.

b. Evaluation of Overall Fuel Recovery Fractions for Shearing LWBR Fuel Rods

Fuel lost during shearing of each individual LWBR fuel rod was determined as the difference between the weight of the unsheared rod and the sum of the weights of the collected segments, including the weight of Zircaloy fines collected on the brushes used for intersegment cleaning of the shear blade (cf. Section IV.D.10). Translation of this calculated weight of lost fuel into a fuel recovery fraction for a rod requires knowledge of the total weight of fuel that the rod contained. Because accurate fuel-stack weights were not available to ANL, it was left for BAPL to calculate the fuel recovery fraction for each rod. We were able, however, to compute approximate values for the fuel recovery fractions, using approximate oxide-fuel weights that BAPL provided on the shearing diagram for each rod. The results of these approximate calculations are shown in Table 14 for the 17 LWBR fuel rods and the performance-verification test rod processed in the campaign. A minimum acceptable recovery of 99.75% had been established for shearing nonirradiated thorium.

As the results in this table show, fuel recovery during shearing was above or very close to the 99.75% target for all the rods except Rod M, the first of five seed rods to be processed at ANL. The lower recovery (99.36%) obtained with Rod M was linked to the use of smaller (3-in. tall by 1.5-in. dia.) aluminum cans to collect the sheared segments as compared to the 6-in. tall by 1.5-in. dia. cans used for collecting segments from the other (larger diameter) rods that were sheared earlier. The shorter cans had been designated for seed-rod segments to provide a consistent concentration of aluminum in the dissolvent when the segments were dissolved; the lower mass of ceramic oxide fuel in the seed-rod segments permitted a lower-volume dissolution. The small cans had

Table 14. Approximate Fuel Recovery for Rods Processed in the EOL Campaign

Rod No.	Rod Type	No. of Cuts	Fuel Lost, g	Approx. Fuel Stack Weight, ^a kg	Approximate Fuel Recovery, %
B	PFB	456	3.23	2.89	99.89
C	PFB	454	3.80	2.86	99.87
D	PFB	454	4.29	2.85	99.85
E	PFB	456	4.46	2.85	99.84
F	PFB	459	2.41	2.88	99.92
G	PFB	453	5.15	2.86	99.82
H	Reflector	287	1.97	7.02	99.97
I	SB	458	3.34	3.42	99.90
J	SB	458	3.37	3.41	99.90
K	SB	456	4.68	3.37	99.86
L	SB	456	2.75	3.41	99.92
M ^b	Seed	733	5.34	0.83	99.36
N	Seed	733	2.20	0.83	99.73
O	Seed	730	2.21	0.83	99.73
P	Seed	731	0.62	0.83	99.93
Q	Seed	732	0.62	0.83	99.92
R	Reflector	286	3.79	6.89	99.95
W ^c	Test	309	1.35	2.12	99.94

^aEstimated from information on rod shearing diagram.

^bSegments collected in 3-in. tall Al cans; others collected in 6-in. tall cans.

^cNonirradiated, PFB-size test rod for performance verification.

adequate volume to collect the longest seed-rod segments, but their use apparently caused greater pressure pulses within the shear's cutting chamber (which includes the can volume) as each cut was made and contributed to a greater carryout (loss) of fine fuel fragments. Because of these results with Rod M, 6-in.-tall cans were used for the remaining seed rods (N through Q), and recovery was markedly improved.

In conclusion, fuel recovery during shearing of the irradiated LWBR fuel rods was satisfactory, meeting in most cases the minimum acceptable recovery limit of 99.75%.

C. Evaluation of Performance Regarding Segment Dissolutions

During the EOL campaign at ANL, 195 dissolver runs were executed over a period of approximately 15 months. These runs included 135 dissolutions of fuel-bearing segments sheared from the LWBR fuel rods; 4 dissolutions of segments

from the mid-campaign performance verification test rod (nonirradiated materials); 17 dissolutions of LWBR fuel rod plenum segments; and 39 blank or cleanup dissolutions to remove traces of dissolved fuel from the dissolver systems after a given rod was processed. Equipment and procedures used for each of these dissolutions were described in Sections III.A.4 and IV.E, respectively.

The segment dissolutions represent a critical first step toward preparing the segment fuel material for analysis. Thus, although no explicit performance criteria regarding the dissolutions were defined in the project error requirements, the dissolver operations had to meet specific demands that were imposed by the requirements on the analytical results. In particular, dissolution of the fuel material had to be complete; the dissolved fuel had to be recovered and accounted for in a way that the analytical results would represent the entire fuel content of the segment; and removal of fuel from the dissolver system had to result in only negligible carryover of fuel material from one dissolution to another. In the first two of the following subsections, we present evidence that the segment dissolutions were complete and the carryover between dissolutions was negligible. In the third subsection, we evaluate fuel losses associated with normal dissolver operations and describe recovery actions that were taken in response to several mishaps that occurred during dissolution of a few segments. The increased uncertainty that was introduced into the analytical results for the segments involved in these mishaps is evaluated in Section V.C.3.b. Because of such mishaps, 5 of the 135 fuel-bearing LWBR rod segments had a potential bias in the segment uranium assay that exceeded its allowable limit; however, all other project requirements were met.

1. Evaluation of Completeness of Segment Dissolutions

Each fuel-bearing segment was taken through the two-stage dissolution procedure described in Section IV.E.3.b. The dissolution completeness was monitored as follows. First, the concentration of uranium in the DS samples (g U/g solution) was multiplied by the weight (usually 1370 g) of reagent (Thorex-A1) used for the second dissolution stage to give an estimate of the total uranium that was left in the dissolver vessel after the primary dissolution stage was finished. This weight of uranium was then expressed as a fraction of the total segment uranium, and the fraction was subtracted from unity to give the estimated fraction of fuel dissolved in the primary contacting with Thorex dissolvent.

Values obtained in this way for the percent dissolution in the primary cycle for segments from the LWBR fuel rods are summarized in Table 15. That the dissolution was complete may be deduced from these data in the following way. The values in the table show that, in general, greater than 90% of the segment fuel was dissolved in the primary cycle. When the dissolvent-to-fuel ratio is high (generally the case for the segments with the highest percent dissolution in the primary cycle), this percentage usually exceeds 99.9%. Thus, it follows that more than 99.9% of the fuel remaining from the primary dissolution period dissolved in the secondary dissolution, giving a percent dissolution for the combined action of the two periods of at least 99.99%.

Additional evidence for complete dissolution of segment fuel comes from observations recorded by the DDS operators during counting and inspection of the

Table 15. Extent of Dissolution in the Primary Dissolution for LWBR Fuel Rod Segments

Rod Designation	No. of Fuel-Bearing Segments Dissolved	% Dissolution in Primary Cycle		
		Lowest	Highest	Average
B	7	89.74	99.97	95.43
C	7	90.74	99.95	97.67
D	7	98.09	99.97	99.76
E	7	99.37	99.97	99.80
F	8	98.68	99.96	99.69
G	7	83.27	99.97	94.26
H	15	99.09	99.98	99.73 ^a
I	8	98.94	99.98	99.69
J	8	99.85	99.99	99.94
K	8	98.18	99.98	99.44
L	8	95.51	99.96	98.67
M	6	98.60	99.96	99.57
N	6	99.64	99.98	99.90
O	6	99.87	99.98	99.95
P	6	98.66	99.98	99.74
Q	6	99.79	99.97	99.93
R	15	99.54	99.94	99.76

^aExcludes data from segment H-08 for which recovery of solution from the primary dissolution was affected by a boilup of solution into the off-gas system (see Section V.C.3).

individual cladding hulls recovered from the tantalum sample basket (see Section IV.E.3.e). In all cases, the operators noted a clean and shiny appearance for these hulls, with no visible traces of undissolved fuel adhering to them.

Another indication of complete segment dissolution is provided by the data from blank dissolutions presented in the subsection below. These data demonstrate a carryover of fuel from one dissolution to the next of less than 0.01%. Fuel carryover in the dissolvers could not have been maintained at these very small levels unless no more than traces of each segment's fuel remained undissolved.

On the basis of the combined strength of the evidence just presented, error (i.e., negative bias) in the ANL segment assay results brought about by incomplete fuel dissolution may be conservatively assigned a limiting value of 0.01%, relative. As we show in Section V.C.4, this error source has a virtually negligible impact on ANL's ability to meet the error requirements on these assays. Hence, in this regard, dissolver performance during the EOL campaign was totally satisfactory.

2. Evaluation of Fuel Carryover Between Segment Dissolutions

Effects of fuel carryover in the dissolvers on the analytical results obtained for dissolved LWBR fuel rod segments were minimized in the ANL operations by (1) sequencing the dissolution of segments from a given rod in order of increasing uranium content and (2) carrying out a "blank" or "cleanup" dissolution in each dissolver system after the last segment from a particular rod was dissolved. The sequencing of dissolutions according to uranium content was established because the uranium assays and isotopic analyses were most sensitive to the potential effects of fuel carryover. From pre-EOL characterization of the dissolver systems, it was known that the amount of uranium carried over between segments was proportional to the uranium in the segment that was dissolved first. Thus, if a high-uranium segment followed a low-uranium segment in the dissolution sequence, the effects of any carried-over fuel would be less important than if the order were reversed. The post-rod cleanup dissolutions were included in the work plan to reduce the potential for carryover from the last-dissolved, high-uranium segments of one rod into the first-dissolved, low-uranium segments of the next.

The extent to which carryover in the dissolver actually occurred is indicated by the results obtained from these post-rod cleanup dissolutions. As with ordinary segment dissolutions, solution from these runs was collected in a clean BT and analyzed for uranium. Results of these blank dissolution runs are presented in Table 16, which shows the uranium recovered in each blank run as a percentage of the uranium in the segment that was dissolved just prior to the blank run. In no case does the uranium found in the blank run exceed 0.01% of the uranium in the previous segment. This level of carryover between segments is acceptably low.

Results given in parentheses for Rods G and Q in this table were obtained from a second cleanup run in the designated dissolver. In these cases, a dual cleanup was deemed desirable to ensure minimal carryover of fuel from the high-uranium segments of these rods into the very low-uranium segments of the reflector rods that were next to be processed. The decrease in uranium recovered during these successive blank runs demonstrates that the blank runs were effective in reducing the level of potential carryover between fuel rods.

3. Evaluation of Fuel Losses Associated with Dissolver Operations

Reliable assay of the uranium and fission products in each LWBR fuel rod segment required not only that the fuel in each segment be completely dissolved, but also that none of the dissolved fuel be left unaccounted for in carrying out the collection and sampling of the resulting solutions. In terms of normal operations with the DDS, complete accounting of the dissolved fuel required that (1) all solutions, except those removed from the system as weighed samples, be completely transferred from the dissolver vessel into the BT; (2) the weights obtained for solution samples from the secondary dissolution and UBT reflect accurately the amount of solution actually removed from the dissolver system [as explained in Section IV.E.3.d.(3), samples from the SBT did not need to be accounted for accurately but were weighed anyway]; and (3) negligible amounts of fuel material be left deposited on the in-line filter through which solution was transferred from the dissolver vessel to the BT. The extent to which each of these operational requirements was satisfied during normal EOL

Table 16. Results of Blank Dissolution Runs
Following Fuel Rod Dissolutions

Rod Designation	% Carryover from DS1 Blank Run	% Carryover from DS2 Blank Run
B	0.0011	0.0100
C	0.0006	0.0036
D	0.0063	0.0100
E	0.0014	0.0100
F	0.0013	0.0024
G	0.0034	0.0099
	--	(0.0025) ^a
H	0.0068	0.0043
I	0.0065	0.0008
J	0.0006	0.0069
K	0.0046	0.0057
L	0.0023	0.0032
M	0.0022	0.0021
N	0.0043	0.0015
O	0.0056	0.0012
P	0.0035	0.0040
Q	0.0014	0.0030
	(0.0008)	(0.0005) ^a
R	0.0075	0.0013
W	0.0083	0.0048

^aResults in parentheses for Rods G and Q were obtained from a second cleanup run in the designated dissolver; a dual cleanup was deemed desirable to ensure minimal carryover of fuel from these rods into the reflector rod segments that were to be processed next.

operations with the DDS is given in Section V.C.3.a, where the amounts of potentially unaccounted for (i.e., lost) fuel associated with each pertinent facet of the operations are estimated. The resulting estimates demonstrate that, although fuel retained on the filter and fuel contained in solution that was routinely lost during sampling may not have been negligible, fuel losses during normal operation with the DDS were small enough to easily meet the limits to allowable error in the segment assays.

Most of the segment dissolutions performed during the EOL campaign were accomplished under these "normal" conditions. However, operational mishaps occurred during dissolutions involving 19 of the 156 segments that were dissolved, including 18 of 152 LWBR fuel rod segments and one segment from the performance-verification test rod. Many of these mishaps were relatively innocuous in that their occurrence affected neither the magnitude of the assay results obtained for the corresponding segment nor the uncertainty associated with the results. The mishaps with eight of the LWBR fuel rod segments, however,

involved a displacement of dissolver solution out of the dissolver system and required special recovery operations to account for the fuel in these displaced solutions. Our approach to dealing with these mishaps was to collect, as completely as practical, the displaced solution, to assay the recovered material, to include these assay results as part of the segment inventory, and to include additional terms in the quoted uncertainty in the assay result that account for the additional measurements and also account for potentially unrecovered material. The effects of these mishaps on the validity of the ANL assay results for the segments that were involved are discussed in Section V.C.3.b. The added uncertainty in these assay results, caused by the potentially incomplete recovery of displaced fuel, raised the potential bias in the segment uranium assay results beyond the limits to allowable error in five cases.

a. Estimation of Fuel Losses Associated with Normal Dissolver Operations

(1) Losses Due to Fuel Left in Dissolver Piping and on In-Line Filter

Given the demonstrated abilities of the ANL dissolvers to attain complete (greater than 99.99%) dissolution of segment fuel material and very small (less than 0.01%) carryover of dissolved fuel in the system, losses of fuel by retention of solution in the dissolver vessel, piping, etc., may be presumed not to have exceeded 0.01%, relative to the amount of fuel in the dissolved segment. Besides these losses, which relate to the transfer of solution from the dissolver vessel to the BT, there are only two other pathways by which fuel in the dissolver vessel can be diverted from collection in the BT in the absence of unusual circumstances: (1) displacement of dissolver solution to the off-gas collection system (mainly the large off-gas hold tank) associated with each dissolver system and (2) deposition of fuel components on the filter in the transfer line by which solution is moved to the BT.

Solution entrained with the off-gas is hindered from entering the gas reservoir by the presence of a 500-cm³ expansion volume (demister) located in the line that connects the dissolver vessel and the tank. Furthermore, this line is plumbed such that solution released from the gas stream in the demister drains back to the dissolver vessel. No evidence of significant amounts of fuel material having left the dissolver vessel by this pathway during normal operations has ever been found, even though recovery operations associated with mishaps that involved inadvertent displacement of solution to the off-gas system provided several opportunities to find such evidence. Radiation surveys of the off-gas-system piping and disassembly and rinsing of the lines following such mishaps showed either no fuel or only the presence of fuel related to the solution that was suspected of being displaced. Hence, losses of fuel through this pathway under normal conditions are considered to have been negligible.

Fuel losses to the in-line solution filter were minimized by two methods. First, the filter housing contained a fine-mesh stainless steel screen in its inlet to prevent all but the finest particles of undissolved fuel from reaching the actual filter medium. Second, the filter and screen were backwashed into the dissolver vessel during addition of reagent for the second dissolution cycle. Fuel retained on the filter during dissolution of nonirradiated material was shown by extensive testing done prior to the EOL campaign to correspond to less than 0.005% of the fuel in the segment that was being dissolved (see, for example,

Ref. 14). The ANL procedures for the EOL campaign did not, therefore, include explicit provisions for determining the amount of fuel retained on each filter. Fuel retention on the filters was monitored, however, on an ongoing basis by measuring radiation levels for the filter media that were used with each EOL segment (see Section IV.E.3.e). Radiation readings obtained for each filter are shown in Table 17.

Table 17. Radiation Levels Measured for Filters Used in Dissolution of Fuel Rod Segments

Segment No.	Radiation Level, R/h																		
	B	C	D	E	F	G	H	I	J	K	L	M	N	O	P	Q	R	W	
00	7	10	10	7	7	5	^a	4	9	5	5	3	5	6	2	5	1	-	
01	8	8	8	8	6	45	4	4	13	7	9	2	6	8	5	6	4	7	
02	70	9	50	30	18	120	5	15	80	25	29	21	11	10	7	25	1	16	
03	120	50	50	45	27	120	6	80	100	30	34	13	50	22	20	25	2	7	
04	120	72	100	50	52	120	6	120	110	35	85	31	60	36	20	12	3	9	
05	120	62	90	17	37	10	30	120	100	80	50	34	8	26	25	7	-	-	
06	40	25	16	40	10	6	15	70	50	55	10	2	7	^a	5	7	4	-	
07	8	8	8	6	7	5	28	100	25	15	10	-	-	-	-	-	3	-	
08	-	-	-	-	7	-	16	60	15	10	8	-	-	-	-	-	20	-	
09	-	-	-	-	-	-	15	4	-	-	-	-	-	-	-	-	4	-	
10	-	-	-	-	-	-	20	-	-	-	-	-	-	-	-	-	6	-	
11	-	-	-	-	-	-	13	-	-	-	-	-	-	-	-	-	20	-	
12	-	-	-	-	-	-	23	-	-	-	-	-	-	-	-	-	10	-	
13	-	-	-	-	-	-	6	-	-	-	-	-	-	-	-	-	2	-	
14	-	-	-	-	-	-	4	-	-	-	-	-	-	-	-	-	4	-	
15	-	-	-	-	-	-	5	-	-	-	-	-	-	-	-	-	2	-	

^aValue for the filter from this segment was not recorded.

During the dissolutions of the most highly irradiated segments of Rods B and C (the first LWBR fuel rods processed at ANL), slow transfers of the dissolver solutions to the BT were encountered, suggesting a possible buildup of material on the filter and partial plugging. These difficulties were alleviated in subsequent dissolutions by increasing the amount of Thorex reagent used in the primary dissolution stage. To evaluate the validity of our assumption that negligible amounts of fuel were being retained on the filter, the filter used in operations with segment B-04 (a very high radiation level, 120 R/h) was examined by gamma spectrometry. The primary source of radioactivity on this filter was Sb-125 and Nb-95. However, some mixed fission products were also found. Among these were Cs-137, Ce-144, and Zr-95 as shown in Table 18. The results for Cs-137 and Ce-144 were taken as an indication that a trace of fuel material was present on the filter, since the two fission products were present in approximately the same proportions on the filter and in the BT solution used for determining the segment totals. The Zr-95 isotope was enriched on the filter relative to the solution, suggesting that most of the material on the filter came

Table 18. Fission Products Found on Filter from Dissolution of Segment B-04

Nuclide	Atoms Found on Filter B-04-FR-1	Total Atoms in Segment B-04	Fraction on Filter, %
Cs-137	2.3×10^{17}	1.66×10^{21}	0.014
Ce-144	1.1×10^{16}	8.07×10^{19}	0.013
Zr-95	6.4×10^{14}	5.88×10^{17}	0.109

from cladding rather than fuel. If the Cs-137 and Ce-144 results are taken as a measure of the fraction of the segment fuel that was lost to the filter during the dissolver operations, one finds that, for this worst-case segment, such losses correspond to only about 0.015% of the segment fuel. Losses of this magnitude do not jeopardize ANL's ability to meet the requirements of the EOL campaign.

Examination of the data in Table 17 indicates that the radiation levels for all rods were consistent with those obtained for segments from Rod B. On the basis of this consistency, we assign a limit to the magnitude of probable fuel losses to the in-line filter of 0.02% of the segment fuel content. On combining this value with the limit (0.01%) to potential fuel losses from incomplete segment dissolution and holdup of fuel in the dissolver system, we conclude that fuel losses associated with the collection of dissolved fuel in the BT under normal operating conditions contributed a negative bias of no more than about 0.03%, relative, to the segment assay results. This bias contribution from the dissolver operations is sufficiently small that it did not jeopardize ANL's meeting the error requirements on these assays; for reference, the requirements permitted a bias of 0.15%, relative, even for the most stringent case, i.e., the total segment uranium assay.

(2) Losses Associated with Dissolver and Blend Tank Sampling Operations

We accounted for solution samples removed from the dissolver systems during the DDS operations by weighing the empty and then the full sample bottles to the nearest 0.001 g, using one or the other of the 300-g capacity balances in the DDS hot cell, and then computing the net weight of solution in each sample from these weight data. The total amount of solution removed as samples from each type of dissolver solution was determined as the sum of the net solution weights in the individual sample bottles. For this sum to provide a valid measure of the weight of solution actually withdrawn from the dissolver vessel or BT, any amount of solution that was removed from the DDS during sampling, but that did not end up in one of the weighed sample bottles, had to be negligibly small. Under normal operating conditions, such solution corresponded particularly to that retained in the disposable sampling lines through which the dissolver solutions were transferred from the system to the sample bottles. The sampling device (see Section III.A.4.d) used for these operations was designed to return solution in the sampling lines to the dissolver or BT and, thereby, minimize the retention of solution in these lines.

The effectiveness of this means for minimizing solution losses during sampling could be checked by the BT weighing operations that were performed as part of the BT sampling procedure [see Section IV.E.3.d.(1)]. The change in weight of the BT assembly upon removal of the UBT solution samples was routinely compared with the sum of the net solution weights for the corresponding sample bottles. Any difference observed between these two independent methods for determining the amount of solution removed from the UBT as samples was ascribed to solution loss during the sampling operations. In general, the change in the weight of the BT assembly was found to slightly exceed the net weight of solution in the sample bottles; solution losses estimated from these data ranged from -1 to +2 g. Because these differences are based on measurements using the BT balances, which had a resolution of only 1 g, their significance was difficult to ascertain. Thus, we did not apply any bias corrections to the segment assay results in accounting for the losses estimated in this way during normal operations. Instead, we included a variance term in our estimates of the uncertainties associated with these results. This term was evaluated as the mean-square magnitude of the losses found for each segment of an individual fuel rod.

This variance (σ_{losses}^2) was combined with the variance associated with the weighings of the sample bottles ($\sigma_{\text{weighing}}^2$), estimated from repetitive weighings performed on each bottle, and the variance associated with calibration of the appropriate balances ($\sigma_{\text{calibration}}^2$), estimated from a comparison of empty-bottle weights obtained inside and outside the DDS hot cell. Thus, an estimate of the total variance associated with the measured weight of solution removed as samples could be obtained from

$$\sigma_{\text{Wt. Sples}}^2 = \sigma_{\text{weighing}}^2 + \sigma_{\text{calibration}}^2 + \sigma_{\text{losses}}^2 \quad (\text{V-7})$$

Each of these terms was automatically evaluated by the report-generating programs used to compute the results of segment assays from a given fuel rod; the value for each term was obtained by statistical analysis or other appropriate calculation using the data recorded during dissolution of segments from the rod under consideration. The magnitude of σ_{losses}^2 was evaluated using data from the UBT sampling operations but was assumed to apply also to the second dissolution samples in subsequent calculations.

The values that were used for these individual terms in calculating the uncertainties reported with the segment assay results for each rod are shown in Table 19. The total variance in the solution sample weights corresponds to a standard deviation of no more than 1.5 g. For typical solution weights of 70 g for the DS samples and 140 g for the UBT samples, this standard deviation translates to a relative uncertainty of 2.14% or 1.07%, respectively. Since the DS samples contained only about 0.1% of the total segment fuel (see Section B.4 of Appendix D), this uncertainty contributes an uncertainty to the segment assay results of only 0.0021% and is virtually negligible. The UBT solution samples typically contained 2 to 3% of the segment fuel, so that the 1.07% uncertainty in the solution weight for these samples translates to an uncertainty in the segment assays of 0.03% (see Section E.2 of Appendix D for more detail regarding this propagation of errors). Although

Table 19. Contributions to Uncertainty in Measured Weight of Solution Removed from the Dissolver System As Samples (Values Estimated from Data Obtained for Segments from Each Individual Rod)

Rod Designation	No. of Segments Used in Making Estimates	Standard Deviation, g			Total Standard Deviation of Weight of Sampled Solution, g
		Individual Weighing	Balance Calibration	Sampling Losses	
B	7	0.0009	0.0045	0.9600	0.9600
C	7	0.0007	0.0026	0.9859	0.9859
D	7	0.0009	0.0044	1.0716	1.0716
E	7	0.0008	0.0039	0.9438	0.9438
F	8	0.0009	0.0045	1.4576	1.4576
G	7	0.0008	0.0043	0.8935	0.8935
H	16 ^a	0.0009	0.0049	0.7236	0.7236
I	8	0.0010	0.0045	0.9502	0.9502
J	8	0.0008	0.0045	1.1333	1.1333
K	8	0.0013	0.0048	1.0303	1.0303
L	8	0.0010	0.0048	1.2215	1.2215
M	6	0.0009	0.0061	0.9614	0.9614
N	6	0.0013	0.0067	0.8851	0.8851
O	6	0.0007	0.0075	0.9964	0.9964
P	6	0.0011	0.0041	0.6077	0.6077
Q	6	0.0012	0.0055	1.1584	1.1584
R	15	0.0009	0.0049	0.9421	0.9421
W	4	0.0009	0.0049	0.9743	0.9743

^aIncludes one dummy segment (H-18) involved in recovery from mishap during dissolution of Segment H-08.

the uncertainty contribution from this error source is important, it is sufficiently small to permit meeting the project error requirements on the assay results.

b. Recovery from Operational Mishaps and Estimation of Associated Fuel Losses

Mishaps that occurred during the EOL segment dissolutions were individually described in the reports to BAPL covering the ANL destructive assay of each fuel rod. The detailed descriptions presented in those reports are not reproduced here. Rather, a summary of the types of mishaps that occurred is presented, and the manner in which mishaps of a given type were handled is illustrated. For mishaps that involved a displacement of fuel-bearing solution from the dissolver systems, we estimated the quantity of fuel that might have gone unaccounted for during recovery operations that followed each mishap. In these estimates, the maximum probable quantity of unrecovered fuel for each segment was established using conservative models to relate process data for these

and similar segments or to define the degree of dispersal of the displaced dissolver solution. These quantities of fuel were treated as contributing to a potential bias in the assay results for the affected segments. These estimates of potential bias are listed for the appropriate segments at the end of the following discussion.

(1) Minor Mishaps during Segment Dissolutions

Many of the mishaps that occurred during segment dissolutions in the EOL campaign were classified as minor mishaps because recovery from them was complete enough that their occurrence was not considered to have introduced significant error into the ANL assay results. A brief discussion of these minor mishaps follows.

During dissolution of segments C-04, D-00, D-02, D-07, and W-03, vapor leaks were observed at the dissolver's gasketed closure. These leaks were stopped by simply tightening the dissolver closure bolts. Subsequent inspection of the gasket area, by means of radiation surveys and analysis of tissue swipes taken to absorb liquid condensed on the dissolver flanges, revealed less than 1 mg of uranium in each case, indicating negligible loss of fuel. For dissolutions after Rod D, we apparently eliminated this problem by retightening the closure bolts during heatup of the dissolver on a routine basis.

During the processing of segments D-02, F-04, and R-08, while the blend tank was being sampled before spiking it with the NBS-950a uranium standard, small amounts (less than 100 g in each case) of the BT solution were spilled. In each case, the weight of solution lost was determined from the measured BT weight change during sampling. Thus, the lost fuel was accounted for in the calculation of the segment assay results. Since the uncertainty reported for each assay result included a term that accounted for errors in this BT weight change regardless of whether or not solution was spilled (see previous subsection), using this information in making the correction for lost fuel did not increase the uncertainty associated with the results.

During the primary dissolution of segment F-03, a malfunctioning pressure-control valve caused depressurization of the dissolver from 118 to 100 psig. Because there was concern that flash-boiling may have occurred and caused transfer of dissolver solution into the off-gas system, the following was done. The line between the dissolver and demister was surveyed for radiation, as were the lower portions of the demister and hold tank. The line was then disconnected from the demister and reverse-flushed with dilute nitric acid, thereby returning any solution in the lines back to the dissolver vessel. Radiation surveys made before and after flushing gave readings near background (1 to 1.5 R/h), indicating that no significant displacement of dissolver solution to the off-gas system had occurred. The pressure-control valve was refurbished to permit the F-03 segment dissolution to be completed. Afterwards, the valve was replaced.

During processing of segment F-08, after the NBS-950a uranium-standard spike was added to the BT, the temperature controller on the BT heater failed, and the BT was overheated. The operator did not notice the malfunction until several hundred grams of the SBT solution had evaporated. It was judged by ANL that the escaped acid vapor contained no fuel; BAPL concurred in this judgment.

(2) Major Mishaps during Segment Dissolutions

In addition to these so-called minor mishaps, the dissolutions of eight segments (C-03, H-08, J-00, J-08, N-02, N-06, J-03, O-03) were marred by events that did affect the assay results. These were classified as major mishaps, although recovery operations allowed project error requirements to be met in all regards, except for potential bias in the uranium assays for five of the eight segments.

Segment C-03. While solution from the second stage of dissolution for segment C-03 was being transferred from the dissolver to the BT, the in-line solution filter plugged and had to be replaced. Care was taken to drain the plugged filter as completely as practical prior to its removal from the system. As the filter housing was removed from the transfer line, we collected all solution that leaked from the associated piping in clean, shallow plastic trays. Upon disassembly, the filter housing was thoroughly rinsed and the filter media were leached with warm 0.5M nitric acid to recover any fuel-bearing solution that might have been left on them. The leakage solution, the filter-housing rinses, and the filter leachate were separately assayed for uranium content. The uranium found in these samples was added to the segment inventory, and the uncertainties in the segment uranium assay results were amended to include the uncertainty contribution associated with this correction. Because the entire correction amounted to less than 0.06% of the total fuel in this segment, potential bias in the segment assays caused by fuel that might not have been recovered in the operations was judged negligible. The standard deviation quoted for the determination of uranium in this segment was, however, slightly larger than it would have been had the mishap not occurred.

Segment H-08. Another of the major mishaps, that involving segment H-08, consisted of two related events. First, midway in the primary dissolution cycle, a flexible transfer line, lined with Teflon and clad in stainless steel, developed a leak in spite of its having passed the pressure test made in the pre-dissolution operations. Since the leak was small, a plastic tray was placed under the leaking line to collect solution that dripped out of the system, and the primary cycle was allowed to continue to completion. At the end of the primary cycle, after cool down of the dissolver was begun, the leak became worse. It changed from a slow drip of solution to a small, directional spray in a downward direction toward the drip pan located under the dissolver's transfer piping. In response to this worsening of the leak, the dissolver operator depressurized the dissolver vessel by venting it to the off-gas system. This action stopped the transfer-line leak, but also led to the second event: the hot solution in the dissolver vessel flash-boiled as a result of the sudden pressure reduction, and solution was carried to the off-gas hold tank. Solution that had escaped the dissolver system through the leaking lines was collected in a catch tray that was placed under the leak when it was first noticed and in a drip pan on the cell work table. This solution was transferred to sample bottles for subsequent assay along with tissue wipes used for mopping up residual traces of the solutions and for rinsing down the catch tray and drip pan with dilute nitric acid.

Solution in the off-gas system was recovered by first draining the system and then disassembling and rinsing it with nitric acid. This was done with special equipment and procedures that were developed specifically in response

to this mishap. The collected solutions and rinses from the off-gas system were transferred into a separate clean BT where they were mixed, sampled, and spiked in the manner used for BT solutions generated during normal segment dissolutions. These operations were carried out using the OPRDSX computerized procedures, and the resulting solution was assigned a dummy segment designation (H-18). Assay results from segment H-18 and from the leakage samples and wipes were added to the results obtained from the segment H-08 operations to obtain the results that were reported for segment H-08. The isotopic composition of uranium in the solutions collected from the off-gas system and from the leakage was used to verify that these solutions actually did contain fuel from segment H-08 and, further, that they contained no extraneous fuel from other segments. To reduce the chance for recurrence of this situation, the flexible hose sections on both dissolvers were replaced with ordinary stainless steel tubing.

The amount of solution that had entered the off-gas system during the H-08 mishap was estimated from a comparison between the solution weight recovered in the BT from the H-08 primary dissolution and the average of the corresponding weights obtained during dissolution of seven similar Rod H segments. This treatment indicated that 920 ± 32 g (out of a total of ~5300 g) of solution from the primary cycle had been displaced to the off-gas system. A sequential-dilution model was then applied to describe the rinsing and recovery operations that were employed in retrieving this solution, and calculations based on this model provided an estimate of the maximum amount of segment fuel that might have been left unrecovered. In estimating the potential bias in the segment assay results associated with this unrecovered fuel, we assumed that the amount of fuel left unrecovered would follow a uniform probability distribution between zero and this maximum probable loss. The expected value for lost fuel (equal to the magnitude of the potential bias) under this assumption is given by one-half the maximum. This estimate of the potential bias also has associated with it a standard deviation given by the maximum divided by $\sqrt{12}$.

A similar treatment was also applied in estimating the amount of fuel that might have been lost during recovery of the solution that had leaked from the failed transfer line. In this case, the maximum probable fuel loss was estimated on the basis of observations made by the dissolver operators, which suggested that no more than about 25% of the leakage solution could have escaped collection in the catch tray or drip pan. The combined results of the data analysis (i.e., error estimates) associated with the solution leakage and boilup events for segment H-08 introduced a potential bias of 0.10% and a relative standard deviation (over and above that related to measurement uncertainties) of 0.04% into the segment assay results. In spite of these mishap-related contributions, total error in most of these assay results did not exceed the limits allowed by project requirements and, therefore, recovery from the mishap was successful. However, potential bias in the results of the segment uranium assay did slightly exceed the allowed limit, as discussed in Section V.E.

Segment J-00. Another major mishap occurred during dissolution of segment J-00, the plenum segment from Rod J, an SB-type rod. The mishap with this segment was unique in that it began with a potentially innocuous mechanical failure (an O-ring blocking the BT inlet line) but was exacerbated by precipitation, in the dissolver and associated piping, of ferrous-alloy plenum components. The precipitate in the dissolver vessel was ultimately

redissolved by reheating the solution, but removal of solids from the transfer piping required disassembly and rinsing of individual transfer lines. Not only was dissolver solution lost in these operations, but extraneous fuel material was also introduced into the system. The presence of such extraneous fuel was inferred, and its quantity was estimated, from uranium isotopic data obtained for samples taken before and after the cleanup operations. Because the plenum dissolutions were performed to screen each rod's plenum for the presence of significant amounts of fuel, and because fuel in the Rod J plenum proved to be negligible, this mishap did not actually affect the validity of the Rod J assay, even though it strongly affected the results for segment J-00. Following this mishap, the use of larger dissolvent volumes in processing plenum segments from similar rods was initiated to prevent precipitation if similar circumstances arose. The EOL campaign was completed without recurrence of a mishap of this type.

Segments J-08, N-02, and N-06. For three segments (J-08, N-02, and N-06), the mishaps involved small leaks in the transfer line that connected the dissolver vessel and BT. In each case, the leak developed at the fitting where the rigid stainless steel tubing section from the dissolver drain line connects to the main shutoff valve on dissolver DS1; this tubing section is the one that replaced the flexible hose after the mishap with segment H-08 (see above). The corresponding line on the other dissolver remained leak-free throughout its use in the EOL campaign.

The leaks that were encountered during these dissolutions were all quite small, involving a drop of solution falling from the fitting now and then over the course of one or both of the dissolution stages. In each case, the leakage solution was collected by placing a plastic catch tray under the fitting and by mopping and rinsing the drip pan that covered the dissolver work-station floor. The collected solution was assayed in each case, and these assay results were included with the corresponding segment inventories. The maximum probable amount of lost fuel associated with each mishap was estimated as a conservatively large fraction of the fuel that corresponded to the uranium present in the recovered solution. We then estimated contributions to the bias and standard deviation of the assay results on the basis of this maximum probable loss, using a treatment analogous to that described earlier for segment H-08. In the worst case (N-06), these potential bias and standard deviation contributions amounted to 0.063% and 0.037% of the assay values, respectively. Thus, because the leaks that defined these mishaps were small, and appropriate recovery actions were taken in a timely manner by the dissolver operators to deal with them, their overall effect on the assay results for the segments involved was slight. Nevertheless, occurrence of the mishaps increased the potential bias in the uranium assay results beyond the limits allowed by the project error requirements for segments J-08 and N-06.

Segment J-03. Operator error in using the interactive process-control (IPC) program during dissolution of segment J-03 resulted in the following mishap. During the primary dissolution cycle, the operator intended to increase the dissolver pressure slightly, to conform to specifications, by adding 125 psig air. Instead, he vented the hot dissolver to the off-gas system when he accidentally and momentarily entered the wrong IPC command number (8 instead of 6) at the computer terminal. The dissolver solution flash-boiled and some solution escaped to the off-gas hold tank. Following the subsequent dissolution of

segment J-04 (which completed the Rod J dissolutions in the DS2 dissolver), draining and rinsing of the DS2 off-gas system were carried out in a manner similar to that used to recover solution from the H-08 mishap. The dissolver solution displaced by the flash-boil during dissolution of segment J-03 was recovered from the off-gas hold tank and piping and was identified as to its source by the isotopic composition of its uranium content.

Although the actions taken to recover the displaced J-03 dissolver solution and its associated fuel material were analogous to those used for the H-08 boilup, they were applied in a way that provided a more thorough rinsing of the off-gas system and recovery of fuel material. As a result, a small amount (3.8 mg) of uranium that was attributable to segment H-08 was recovered in addition to the J-03 fuel materials. The identity of this H-08 uranium was inferred from its effect on the isotopic composition of the recovered J-03 uranium, and its amount was estimated from conventional isotope dilution formulas. The assay results for segment H-08 were subsequently amended to include this amount of uranium. Similarly, the assay results for segment J-03 were also amended to include the results of assays performed on solutions recovered from the off-gas system.

In assigning uncertainties to the J-03 assay results, we conservatively assumed that the maximum amount of fuel that might have been left unrecovered in the off-gas system was of the same magnitude as that recovered during the rinsing operations. In a similar manner to the treatment used in handling data from the other mishaps described earlier, we assumed that the amount of fuel that was actually left unrecovered followed a uniform distribution between zero and that maximum value. The potential bias and standard deviation associated with this unrecovered fuel in the case of the J-03 segment corresponded to 0.117% and 0.067%, respectively, of the total segment fuel. Even with these conservative estimates of error associated with incomplete recovery of the displaced solution, total uncertainty in the J-03 assay results were found to meet project requirements in every case except that of potential bias in the uranium assays. The limit to allowable bias in the uranium assays was slightly exceeded, as discussed in Section V.E.

Segment O-03. Another operator error in using the IPC program, which occurred in spite of preventive actions taken to avert such errors following the J-03 mishap, resulted in a serious mishap during dissolution of segment O-03. In this case, an operator planned to evacuate the vacuum reservoir tank for DS2; but, because of incomplete IPC entry (1 instead of 19), he configured the system for solution addition to the dissolver through the spray ring while the primary dissolution was in progress. Under this condition, the dissolver vessel vents to the hot cell to allow escape of air displaced by the incoming reagent. Pressure in the hot dissolver was thus vented to the cell; the solution boiled up owing to the rapid depressurization; and some solution was carried out of the dissolver with escaping vapor. Unlike the boilups that occurred for segments H-08 and J-03, for which the solution displaced from the dissolver entered a closed system (i.e., the off-gas system) and was retained in a location where it could be recovered with some confidence, this mishap with segment O-03 displaced the solution into the open cell, where the effectiveness of recovery operations was more difficult to ascertain. Physical evidence, from radiation surveys of the cell and from consideration of the trajectory of the steam plume that left the dissolver when it was vented, guided the recovery operations and

indicated that the dispersal of solution in the cell was not widespread. Fuel from the displaced solution was recovered as completely as practical by repeatedly wiping down and rinsing the cell's work table, equipment, walls, and windows in the areas where the spray was directed. Assays performed on the wipes and rinses generated during these cleanup operations indicated that the amount of fuel that had left the dissolver was relatively small (approximately 0.5% of the segment total) and confirmed that fuel dispersal was not widespread. Most of the fuel material was found in wipes from a small area adjacent to the outlet of the dissolver vent line. The material recovered through these operations was verified as belonging to segment O-03 by its uranium isotopy and was included in the segment inventory.

Completeness of recovery was ultimately assessed by means of a radiation survey of the affected areas in the cell. Because the sensitivity of the in-cell radiation probe was limited, however, we could confirm only that the amount of fuel left unrecovered did not exceed an amount equal to about 0.44% of that present in segment O-03. Using this value as the maximum probable fuel loss associated with the segment O-03 mishap and applying a data treatment similar to that described earlier, we concluded that potentially unrecovered fuel associated with the O-03 mishap contributed a potential bias to the assay results of 0.22%, relative, and a relative standard deviation of 0.13%. This bias contribution exceeds, by itself, the limit (0.15%) to allowable bias in the uranium assay for an individual segment as prescribed by the project error requirements. Thus, this requirement was not met for segment O-03 as a result of the mishap that occurred during its dissolution. However, the estimated standard deviation of the uranium assay result for this segment was below the limit to allowable error of 0.15%. All other assay results, i.e., those for the fission-product monitors, met both the requirements on bias and standard deviation.

4. Conclusions Regarding Performance During Segment Dissolution

The foregoing discussions demonstrate that, under normal conditions, fuel losses associated with segment dissolutions during the EOL campaign were limited to about 0.03% of the segment fuel loading, a small enough fraction to meet the error requirements on the individual segment assays. Even for those segments that were involved in operational mishaps that affected the dissolver operations, recovery of fuel materials was sufficient (see summary of estimated losses in Table 20) to meet most requirements.

Considering that 156 segments were dissolved over the course of the 15-month EOL campaign, and that these dissolutions were carried out on a grueling 24-hour-per-day schedule involving some 14 different dissolver operators, we believe that the level of performance achieved in these operations is extraordinarily good. A large share of the credit for the success of the EOL dissolver operations is attributable to the use of fully computerized procedures, which provided nearly exact replication of the processing sequence for each segment and improved the operator's ability to monitor process equipment for signs of actual or impending problems. Nevertheless, the role of the operators in achieving this high level of success warrants special recognition. Without their vigilance toward recognizing off-normal situations and their promptness and care in dealing with such situations when they occurred, this level of performance could not have been achieved, much less maintained over so long a period of time.

Table 20. Summary of Estimated Fractional Fuel Losses During Dissolver Operations

Segment Designation	Estimate of Potentially Unrecovered Fuel, ^a %	Uncertainty in Estimate, %
H-08	0.13 ^b	0.04
J-03	0.15 ^b	0.07
J-08	0.07 ^b	0.02
N-02	0.03	0.005
N-06	0.09 ^b	0.04
O-03	0.25 ^b	0.13
All Others	0.03%, max.	--

^aCalculated as the sum of losses (taken as 0.03% of segment fuel) under normal operations and loss due to occurrence of dissolution mishap.

^bResulted in failure to meet requirement on bias in segment uranium assay.

D. Precision and Accuracy of Uranium Isotope Abundance Determinations

The isotopic composition of uranium from each dissolved segment was determined by analysis of single aliquots taken from each of two separate samples of the UBT solution. The weight percent abundance of each uranium isotope was calculated from measured ratios of the individual isotopes to ²³³U according to the relationships

$$WPC_i = \frac{100\%}{1 + \sum_j R_j}, i = 233 \quad (V-8a)$$

$$WPC_i = \frac{100\% R_i}{1 + \sum_j R_j}, i = 232, 234, 235, 236, 238 \quad (V-8b)$$

Here, WPC_i stands for the weight percent abundance of isotope i ; R_i stands for the measured weight ratio ${}^i\text{U}/{}^{233}\text{U}$; and $\sum R_j$ is the sum of measured ratios for all isotopes (other than ²³³U) present in the segment uranium. For ²³²U, this ratio was measured by Alpha Pulse Height Analysis (APHA), as described in Section IV.F.1.d.(2). For the other isotopes, the ratios were measured by thermal ionization mass spectrometry (TIMS) as discussed in Section IV.F.1.d.(1).

The weight percent abundances were calculated using a computer program that not only compiled the APHA and TIMS data required by the calculation but also carried out a statistical analysis of the ratio data and propagated the resulting error estimates to derive an estimate of the uncertainty

associated with each reported isotope abundance. The uncertainty quoted for each result represented an estimate of the standard deviation of the reported value and included contributions from randomly varying systematic errors (i.e., errors that affected all results from a given segment to about the same extent but varied randomly among segments), as well as from random errors that could be evaluated from observed differences between replicate measurements that were performed for each isotope of each segment. Potential bias errors in the EOL results were not explicitly quoted in the rod reports.

Although weight percent abundances of all uranium isotopes (mass numbers 232, 233, 234, 235, 236, and 238) were reported for each segment, the fissile isotopes (^{233}U and ^{235}U) were of primary interest to BAPL. Project error requirements permit a potential bias in the sum of the abundances of these isotopes of 0.05% and a standard deviation of the sum of 0.08%, relative. (For segments of low uranium loading, i.e., less than 0.2 g total U in the segment, these requirements were relaxed somewhat; cf. Table 2.)

Estimation of the potential bias and standard deviation for each weight percent abundance determined by ANL is treated in detail in Appendix B. Bias errors in these determinations arose from two sources: uncertainty in the NBS isotopic Standard Reference Material (SRM), U-500, used in calibrating the TIMS isotope ratio measurements, and error in the ^{232}U half-life used in computing R_{232} from APHA data. The magnitude of the percent relative bias associated with each individual weight percent abundance depends on the specific isotopic composition of the uranium sample. However, by assuming a worst-case hypothetical composition consistent with the isotopy of the uranium in the LWBR fuel rod segment, we show (in Appendix B) that the maximum probable bias in the sum of the weight percent abundances of the fissile isotopes is only 0.016%, relative, and is well within the limit (0.05%) to allowable bias specified in the project error requirements. No requirements were established for determination of the other (i.e., non-fissile) uranium isotopes. Estimates of the maximum probable bias in the weight percent abundances of each of these isotopes is, nevertheless, presented in Table 21 for completeness. We note that the percent relative bias in the sum of ^{233}U and ^{235}U is slightly smaller than the weighted sum of the individual biases because of a partial cancellation of errors when the sum is taken.

Random error sources that were considered in estimating the standard deviations quoted with each uranium isotopic result included the calibrations of measurement systems, the measurements themselves, and the introduction of traces of extraneous uranium into the samples prepared for analysis (i.e., the isotopic procedure blank). Individual error contributions from each of these sources are also discussed in detail in Appendix B, where we use EOL data from both samples and standards to support the validity of our approach to assigning these limits to the probable random error in our results. The percent RSD of the sum of the fissile uranium isotopic abundances for each individual segment processed during the EOL campaign is shown in Table B-3 of Appendix B. Every segment analysis met requirements on the standard deviation of this sum. In fact, except for two low-uranium segments (D-01 and R-01), which showed standard deviations between 0.08 and 0.09%, relative, the RSD of the sum never exceeded 0.03%. Precision of the EOL isotopic measurements was, thus, eminently satisfactory.

Table 21. Estimates of Maximum Probable Bias in Weight Percent Abundances Reported for Individual Uranium Isotopes

Mass Number of U Isotope	Estimated Maximum Potential Bias, % Relative
232	+4.0
233	± 0.019
234	± 0.022
235	± 0.038
236	± 0.055
238	± 0.088

To provide an ongoing check on the quality of uranium isotopic measurements made during the EOL campaign, we analyzed the UNH (uranium nitrate hexahydrate) reference material that was provided by BAPL for this purpose. Results of these analyses are reported in Table 22. The UNH material was analyzed by TIMS at a frequency of at least once per 10 fuel rod segments during the EOL campaign, in accord with the specifications appended to the project error requirements. These results provide a measure of the precision and accuracy of our TIMS measurements only, since the UNH material contained no ^{232}U and was not subjected to isolation and purification chemistry prior to being run. The standard deviations on the individual weight percent abundances shown in Table 22 for the various isotopes correspond to an imprecision in the measured isotope ratios of only 2 to 4 ppm, absolute. This level of imprecision is consistent with the level of random noise associated with our mass-spectrometer measuring systems, as it should be for the small ratios measured in analyzing this reference material. Thus, the results obtained with the UNH reference material are consistent with the error treatment described in Appendix B.

E. Precision and Accuracy of Uranium Assay Results

1. Description of Uranium Assay Methodology

The uranium content of each dissolved segment was determined by combining data from measurements made on samples of solution taken from the dissolver and BT. For fuel-bearing segments, the total segment uranium was calculated in two virtually independent ways.

In one case, the total segment uranium was computed from the quantity of uranium present in the SBT solution by adding to it the (small) quantities of uranium that were removed from the dissolver system in the DS and UBT samples. These latter quantities were determined as the product of the weight of solution present in the samples taken from the dissolver or BT and the weight concentration of uranium in the corresponding solution. Weighing of the DS and UBT solution samples was described in Sections IV.E.3.b.(2) and

Table 22. Data from Individual Analyses of UNH Reference Material

Date	Comp. of UNH Material, wt %				
	233	234	235	236	238
10-10-83	98.4545	1.2158	.0204	.0003	.3091
11-15-83	98.4548	1.2154	.0202	.0004	.3091
11-15-83	98.4536	1.2159	.0203	.0006	.3095
01-03-84	98.4533	1.2156	.0203	.0004	.3103
08-22-84	98.4541	1.2154	.0205	.0006	.3094
08-22-84	98.4526	1.2156	.0207	.0006	.3104
09-25-84	98.4534	1.2160	.0203	.0003	.3099
11-06-84	98.4539	1.2157	.0201	.0005	.3097
11-27-84	98.4524	1.2162	.0205	.0006	.3103
12-13-84	98.4546	1.2155	.0202	.0002	.3094
01-08-85	98.4543	1.2155	.0203	.0003	.3096
02-04-85	98.4538	1.2155	.0206	.0005	.3096
02-27-85	98.4539	1.2156	.0202	.0004	.3099
03-20-85	98.4540	1.2156	.0203	.0004	.3097
04-09-85	98.4549	1.2157	.0199	.0001	.3094
05-21-85	98.4544	1.2154	.0201	.0004	.3096
06-12-85	98.4539	1.2158	.0205	.0001	.3097
07-09-85	98.4537	1.2155	.0204	.0006	.3098
07-22-85	98.4536	1.2155	.0207	.0006	.3097
03-06-85	98.4548	1.2155	.0201	.0003	.3093
09-04-85	98.4536	1.2154	.0207	.0004	.3100
09-16-85	98.4534	1.2161	.0203	.0003	.3099
10-15-85	98.4533	1.2159	.0206	.0002	.3100
10-23-85	98.4545	1.2156	.0202	.0003	.3093
Avg.	98.4539	1.2157	.0204	.0004	.3097
Std. Dev.	.0006	.0002	.0002	.0002	.0004
Reference Values	98.454	1.218	.020	.000	.308

IV.E.3.d(1). The required concentrations were measured by mass spectrometric isotope dilution (MSID) analysis of aliquots weighed from the DS and UBT samples and spiked with NBS 993, as described in Section IV.F.1. Because uranium in the SBT solution represented the bulk (97% or more) of the segment uranium and because it was measured by MSID with the NBS 950a assay standard as spike [see Section IV.E.3.d(2)], the total segment uranium result that was calculated from it was designated the 950a-MSID uranium assay result, ${}^T\text{U}_{\text{seg},950}$. It may be expressed mathematically as

$${}^T\text{U}_{\text{seg},950} = \text{U}_{\text{SBT}} + W_{\text{SDS}} [\text{U}]_{\text{DS}} + W_{\text{UBTS}} [\text{U}]_{\text{UBT}} \quad (\text{V-9})$$

where U_{SBT} stands for the segment uranium in the SBT solution, W_{SDS} is the weight of solution removed from the dissolver as DS samples, W_{UBTS} is the corresponding weight of UBT samples, and $[U]$ represents the weight concentration of uranium in solution from the subscripted source.

The other way in which total uranium was calculated for fuel-bearing segments involved summing the uranium removed from the dissolver as DS samples and the uranium contained in the UMU solution from the blend tank. This solution was weighed during dissolver operations as described in Section IV.E.3.d.(1). The quantity of uranium present in the solution was computed as the product of this weight and the concentration, $[U]_{\text{UBT}}$, that was determined by measurements on the UBT samples, as described above. Because the uranium concentrations involved in this calculation were determined by MSID with NBS 993 as the spike material, the result of the calculation was designated the 993-MSID assay result, ${}^T U_{\text{seg},993}$. Thus, we may write

$${}^T U_{\text{seg},993} = W_{\text{SDS}} [U]_{\text{DS}} + W_{\text{UBT}} [U]_{\text{UBT}} \quad (\text{V-10})$$

where W_{UBT} stands for the UMU solution weight measured during the dissolver operations.

The 993-MSID result was considered to be a corroborative check on the uranium assay for each fuel-bearing segment, supporting the primary assay result obtained from the 950a-MSID calculation. Corroboration of the 950a-MSID results by the 993-MSID results was eminently satisfactory. Percent relative differences between the two results for a given segment averaged only 0.0175% over the 135 fuel-bearing segments processed during the EOL campaign and showed a standard deviation of 0.0889% (see Fig. D-3 of Appendix D). Both the magnitude and dispersion of these differences are consistent with expectations based on a comprehensive analysis of errors associated with each of the two methods. Details of this error analysis are presented in Appendix D, which also includes a more-explicit description of the calculations involved in the MSID measurements. The 993-MSID assay results are not considered further in the present discussion.

In addition to uranium assays on the fuel-bearing segments, we performed similar assays on the plenum segments from the LWBR fuel rods to screen the plenums for the presence of fuel material. Plenum segments were also assayed for thorium to enhance the sensitivity and value of this screening procedure. For these assays, weighed aliquots of UBT solution samples from the plenum dissolution were mixed with a double spike containing both 20 μg of NBS 993 and 100 μg of ${}^{230}\text{Th}$ as described in Section IV.F.1.c.(1). The concentrations of thorium and uranium in the UBT solution were determined by MSID with these spiked aliquots; total U and Th in the plenum were calculated from these concentrations and the UMU weight of the BT solution in a manner analogous to the 993-MSID calculation. No term corresponding to the DS samples (cf. Eq. V-10) was required for the plenums because their dissolution involved only a single contacting with dissolvent and no DS samples were taken (see Section IV.E.3.b).

Results of uranium and thorium assays performed on the plenum segments are summarized in Table 23. These results indicate the presence of fuel material in virtually all plenum segments, although the quantities of fuel involved were negligibly small in most cases. For three (D, E, and K) of the 17 fuel rods, the quantity of uranium in the dissolved plenum was sufficient to determine the uranium isotopic composition. In each of these rods, the isotopy of the plenum uranium was found to agree closely with that of the fuel-bearing segment adjacent to the rod plenum. This result indicates that the origin of this fuel was the end of the fuel stack rather than carryover, on the shear or in the dissolver, of fuel from other rods. Although dissolution and assay of the plenum segments are regarded as having been worthwhile, since in at least one case (K) the fuel that was recovered represented a significant, albeit small, fraction of the fuel in the adjacent fuel-bearing segment, the plenum assay results are ignored in the following discussion of errors associated with ANL's uranium assay results. This is valid because the screening assays were not subject to error requirements, and, owing to the very small amounts of fuel involved, their associated errors are negligible.

Table 23. Summary of Uranium and Thorium Assay Results for Dissolved Plenum Segments

Rod No.	Thorium in Dissolved Plenum, g as ThO ₂	Uranium in Dissolved Plenum, mg
B	0.065 ± 0.000	0.04 ± 0.01
C	0.014 ± 0.000	0.16 ± 0.02
D	0.052 ± 0.000	0.42 ± 0.02 ^a
E	1.007 ± 0.006	0.80 ± 0.02 ^a
F	0.029 ± 0.000	0.06 ± 0.01
G	0.015 ± 0.000	0.11 ± 0.01
H	0.062 ± 0.000	0.05 ± 0.01
I	0.021 ± 0.000	0.11 ± 0.01
J	0.240 ± 0.120	0.15 ± 0.08
K	4.747 ± 0.024	2.36 ± 0.02 ^a
L	0.024 ± 0.000	0.36 ± 0.02
M	0.009 ± 0.000	0.07 ± 0.01
N	0.008 ± 0.000	0.09 ± 0.01
O	0.031 ± 0.000	0.15 ± 0.01
P	0.012 ± 0.000	0.29 ± 0.01
Q	0.007 ± 0.000	0.08 ± 0.01
R	0.007 ± 0.000	0.03 ± 0.01

^aUranium quantity sufficient for isotopic characterization.

2. Bias and Standard Deviation of Segment Uranium Assay Results

Estimation of the potential bias and standard deviation for each segment uranium assay is treated in detail in Appendix D. Bias in the total segment uranium results arose from two primary sources: (1) potential errors in

the certified values of standards used in the measurements associated with the assays and (2) potential losses of fuel material during dissolver operations to prepare each segment for analysis. The maximum-probable measurement bias in assays performed using the 950a-MSID method was 0.093%, relative, as estimated on the basis of the one-sigma uncertainties of the NBS 950a, NBS 993, and NBS U-500 standards that were employed. Under the conditions of normal dissolver operations, fuel losses did not exceed 0.03% of the segment total, as shown in Section V.C. Therefore, by summing the fuel-loss bias and measurement bias for segments dissolved under normal conditions, we determined a limit to probable bias in the 950a-MSID assay results of 0.12%, relative. The uranium assays for these segments thus met the project requirement that this bias be less than 0.15%, relative.

During dissolver operations with eight of the 135 fuel-bearing segments that were processed in the EOL campaign, there were mishaps that increased the potential for losing segment fuel (see Section V.C.3.b). As a result, when the measurement bias and fuel-loss bias were combined for these segments, potential bias in the 950a-MSID results for five of them exceeded the 0.15% limit (see Table D-3 of Appendix D). For the worst of these mishaps, involving segment 0-03, our limit to probable bias in the 950a-MSID result rose to 0.34%, relative.

To evaluate the significance of our having failed to meet project requirements regarding bias error in the uranium assays for these few segments, one must consider these errors in the context of the DOE-NR parametric requirements on which the project requirements were based (Section II.B). If it can be shown that our segment uranium assay data met the relevant DOE-NR parametric requirements despite the larger-than-specified potential for bias error in the assay results for a few segments, then these results may be judged acceptable in terms of achieving overall project goals. The parameters that are of primary interest in regard to judging the quality of our segment uranium assays are the whole-rod loadings of total and fissile uranium, each of which is calculated from the segment assay results and has an allowable bias of 0.15%, relative (see Table 2).

The whole-rod loadings of total and fissile uranium were calculated by BAPL using the sums of the segment assay results and the fuel recovery fraction obtained from ANL's shearing data (Section V.B.3.b). Since the relative bias in a sum of values, each value having the same relative bias, is equal to the relative bias in each individual value, it follows that the purpose of the project error requirement will be fulfilled if the relative bias in the sum of the segment assay results does not exceed 0.15%. In the case where the individual segments may have different relative bias values, relative bias in this sum is given by the expression

$$B_R(^T U_{Rod}) = \frac{1}{^T U_{Rod}} \sum_{seg} ^T U_{seg} B_R(^T U_{seg}) \quad (V-11)$$

Here, the notation $B_R(X)$ represents the percent relative bias in the variable X , $^T U_{Rod}$ stands for the sum of either the total or fissile uranium over segments

from a given rod, and $T_{U_{seg}}$ stands for the uranium loading of individual segments from the same rod. As shown in Section D of Appendix D, our estimates of the maximum-probable relative bias in the fissile loading of each segment is virtually identical to the corresponding bias in the total uranium loading as determined by the 950a-MSID method. Because the distribution of fissile uranium among the segments from each rod is likewise very similar to the distribution of total uranium, values for the relative bias in the sum of the segment assay results for each rod are the same whether total or fissile uranium is considered in applying Eq. V-11.

Results of calculations based on this relationship are shown in Table 24 for each LWBR fuel rod. None of these estimates of maximum probable bias exceeds 0.15%, relative. Thus, the limit (0.15%) to allowable bias error specified in the DOE-NR parametric requirements for the whole-rod assay results are satisfied, even though bias errors in the results for a few of the individual rod segments exceed the limit set by the project requirements. Thus, we feel justified in concluding that recovery from the dissolver-operations mishaps was successful and that, on the whole, ANL's uranium assays were of sufficient quality to meet LWBR-POB project needs.

Also listed in Table 24 are estimates of the standard deviation of the sum of segment assay results for each LWBR fuel rod. These were evaluated using the relationship

$$\sigma_R^2(T_{U_{Rod}}) = \frac{1}{T_{U_{Rod}}^2} \sum_{seg} T_{U_{seg}}^2 \sigma_R^2(T_{U_{seg}}) \quad (V-12)$$

and values of the standard deviation of each individual segment assay, as derived in Appendix D for the 950a-MSID uranium assays. In Eq. V-12, $\sigma_R(X)$ stands for the percent relative standard deviation of the variable X, and the other variables are defined for Eq. V-11.

In estimating the standard deviations of the 950a-MSID assay results, we took into account sources of random error and randomly varying systematic error. Included were errors associated with the preparation of MSID spikes, the taking and weighing of solution samples and aliquots, and the measurement of uranium isotope ratios by mass spectrometry. The standard deviation of each 950a-MSID assay result depended on a number of segment-specific variables and was separately evaluated for each individual segment. Values for these estimated standard deviations ranged from 0.018 to 0.185%, relative, with the value for only one of the 135 fuel-bearing segments exceeding the limit (0.15%) allowed by project error requirements. This segment (C-01) had a low uranium loading (0.22 g) that represented only a minor fraction of the uranium total (58.22 g) for the rod segments. Its larger relative standard deviation (0.185%) was judged to be acceptable since it did not significantly affect the relative standard deviation of the sum of the individual segment uranium assays for this fuel rod. Hence, project requirements in regard to standard deviation of the segment uranium assays were satisfied.

Table 24. Sums of Segment Uranium Assay Results and Estimated Limits to Probable Error for LWBR Fuel Rods

Rod No.	Sum of Total Segment Uranium Assay Results ^a			Sum of Fissile Uranium Assay Results ^a		
	Mass, g	Max. Probable Rel. Bias, %	Rel. Std. Deviation, %	Mass, g	Max. Probable Rel. Bias, %	Rel. Std. Deviation, %
B	56.7030	0.12	0.012	51.4762	0.12	0.012
C	58.2180	0.12	0.012	53.6837	0.12	0.012
D	60.3196	0.12	0.012	57.3257	0.12	0.012
E	31.6046	0.12	0.011	29.2287	0.12	0.011
F	36.2299	0.12	0.013	33.8926	0.12	0.013
G	49.8488	0.12	0.012	45.1988	0.12	0.012
H	36.3039	0.13	0.011	34.7730	0.13	0.011
I	57.7890	0.12	0.010	52.2887	0.12	0.010
J	40.6468	0.14	0.017	36.5979	0.14	0.017
K	51.7439	0.12	0.011	47.1654	0.12	0.011
L	44.3847	0.12	0.012	40.6113	0.12	0.012
M	25.6756	0.12	0.012	22.7935	0.12	0.013
N	28.1974	0.12	0.011	25.5556	0.12	0.012
O	15.6455	0.15	0.022	13.8627	0.15	0.022
P	17.6294	0.12	0.011	15.6404	0.12	0.011
Q	19.6808	0.12	0.014	17.4292	0.12	0.014
R	24.4173	0.12	0.007	23.7248	0.12	0.007

^aNot corrected for fuel losses during shearing.

F. Precision and Accuracy of Fission Product Determinations

The fission products ^{137}Cs , ^{144}Ce , and ^{95}Zr were determined for each fuel-bearing segment by gamma spectrometry on weighed aliquots of the UBT solution samples. A high-purity germanium (HPGe) detector and fully automated multichannel analyzer/data management system were used for these determinations. All fission product data were referred to 12:00 noon on January 1, 1984. The half-lives for the fission products were taken to be 30.17 y for ^{137}Cs , 284.3 d for ^{144}Ce , and 64.02 d for ^{95}Zr .

Details of the fission product determinations are given in Appendix E together with a comprehensive discussion of errors associated with the fission product results. Key observations and conclusions from that discussion, particularly as regards potential bias errors and standard deviations for the results, are summarized below.

Potential bias in the fission product determinations arose from two main sources: (1) potential errors associated with the measurements that were made in the course of each determination and (2) potential losses of fuel material during dissolver operations. As was done for estimating the probable bias in ANL's uranium assay results for each segment (see Section V.E.2), probable bias in the

fission product determinations was estimated by summing contributions from the measurement and fuel-loss biases. Systematic errors in the measurements arose primarily from uncertainties in the certified values of standards used in calibrating the gamma-counting system (see Table E-2 of Appendix E) but also included minor contributions from potential errors in weighing the UBT solution and sample aliquots. Limits to the magnitude of probable fuel losses during dissolution of individual segments are shown in Table 20. Limits to the magnitudes of probable bias errors, calculated as the sums of the fuel-loss and measurement biases for each segment, are given in Table E-4 of Appendix E for all three fission products.

The maximum probable bias values given in Table E-4 are smaller than the limits to allowable bias, as called out in the project error requirements, in every case. For ^{137}Cs , the largest maximum probable bias estimate was encountered for segment 0-03; it had the value 0.43%, relative, well within the allowed limit of 0.5%, relative. For ^{144}Ce and ^{95}Zr , the largest values for these estimates occurred with this same segment and had identical magnitudes, equal to 0.70% in each case. For comparison, the limits to allowable bias in determination of these fission products were 2.0% for ^{144}Ce and 2.5% for ^{95}Zr . Clearly, project requirements regarding bias in the fission product determinations were satisfied.

Precision of the fission product determinations was limited, to a large extent, by counting statistics associated with measuring the appropriate gamma-peak intensities, but it was also influenced by random errors in weighing the sample aliquots and the UBT solution and in calibrating the gamma spectrometer. Additional variance contributions had also to be included for those few segments that were involved in mishaps during dissolver operations. Separate estimates of the relative standard deviation for each fission product determination on each segment were generated by the computer program that compiled and processed the fission product data. These estimates are listed in Tables E-5, E-6, and E-7 of Appendix E for ^{137}Cs , ^{144}Ce , and ^{95}Zr , respectively.

Standard deviations of the ^{137}Cs determinations ranged from 0.25 to 0.56%, well within the 1.25% limit allowed by project requirements. The ^{144}Ce determinations also met requirements (allowable RSD of 2.0%), with estimated RSD values ranging from 0.49 to 1.10%.

For determination of ^{95}Zr , all error requirements were waived for measurements performed more than two years after shutdown of the LWBR and for segments of low burnup or low ^{95}Zr concentration (cf. Footnote b of Table 2). These waiver conditions, which translated to an exemption from requirements of all ^{95}Zr measurements performed after October 1984, were established because ongoing radioactive decay of this short-lived nuclide (half-life of 64.02 d) made measuring it difficult. Only rods B, C, and D were processed prior to the October 1984 waiver date. Among the segments from these rods, the % RSD of the ^{95}Zr determination for only one segment (D-07) failed to meet the 4.0% limit allowed by project requirements. Because this segment had a low concentration of ^{95}Zr , it was exempt from the requirement. Thus, all ^{95}Zr determinations for which the requirement was pertinent showed acceptable levels of precision; RSD values ranged from 1.21 to 3.76%.

We continued to report values for the ^{95}Zr in each segment processed after the waiver date along with our best estimate of the uncertainty associated with the reported value. For segments from the PFB, SB, and first reflector rods, estimated RSD of the ^{95}Zr measurements was generally on the order of 10% or less. For nine of the 45 segments from the seed and second reflector rods, the ^{95}Zr gamma peaks were swamped by counts from ^{154}Eu , which has peaks at the same gamma energies as ^{95}Zr . In these cases, the presence of ^{95}Zr in the segments could not be verified and was reported as "not detectable." Detection limits based on counting statistics associated with the measurements were given in place of uncertainties for these determinations. Other segments from the seed and second reflector rods showed much smaller $^{154}\text{Eu}/^{95}\text{Zr}$ ratios, similar to the other rod types, and provided measurements with RSDs in the range of 3 to 30%. To achieve these levels of precision late in the campaign, we had to extend the counting time (live time) for each ^{95}Zr aliquot to 60,000 s (~17 h), the maximum time our gamma spectrometer could accommodate.

G. Observations Regarding Fission-Gas Measurements

Measurements of the amounts of fission gas (Xe and Kr) contained in each fuel rod plenum and in off-gas accumulated during each segment dissolution were made on a best-effort basis by gas mass spectrometry. When combined with the estimated mass of fission gas released during rod shearing, as measured with the in-line radiation monitor in the shear cell ventilation system (see Section III.A.2.c), these data provided a clear view of the distribution of fission gas within each fuel rod. A summary of fission-gas distribution for rods B through Q is presented in Table 25. Corresponding data for rod R, the last rod processed in the EOL campaign, are not available because off-gas samples from the dissolution of segments from this rod were not taken (see Section IV.E.3.c). As the table shows, virtually all (more than 99%) of the fission gas in each rod remained with the fuel material until it was released by dissolution of the fuel.

On a few occasions during processing of the LWBR fuel rods, dissolver off-gas samples were lost owing to such problems as pump failures and leaks in the off-gas retention system. Rather than report no value for the fission gas contained in these segments, we reported values that were estimated by means of an apparent correlation that existed between the fission-gas content and the ^{137}Cs content of segments belonging to a given rod. It was interesting to find that a single correlation line could be applied to all rods processed in the EOL campaign, as illustrated in Fig. 31, which plots fission gas vs. ^{137}Cs for each of the LWBR fuel rod segments. The plus and minus 10% limits on the plot represent the estimated one-sigma uncertainty of the fission-gas measurements. The consistency of the fission-gas results relative to the ^{137}Cs determinations supports the validity of the fission-gas values and the error limits that we assigned them.

H. Conclusions

The ultimate aim of the LWBR-POB Analytical Support Project at ANL was to provide high-quality physical and chemical data on the EOL fuel rods that we analyzed destructively and to accomplish the destructive assays in a timely manner. Years of effort (Phases I and II of the project) were expended in

Table 25. Distribution of Fission Gas Released during LWBR Fuel Rod Processing

Rod No.	Fission Gas Released during Specified Operations, wt %		
	Plenum Puncture ^a	Shearing ^b	Dissolution ^c
B	0.07	0.26	99.67
C	0.03	0.28	99.69
D	0.01	0.28	99.71
E	0.02	0.23	99.76
F	0.02	0.18	99.80
G	0.06	0.39	99.55
H	0.01	0.17	99.82
I	0.05	0.23	99.72
J	0.03	0.31	99.66
K	0.03	0.17	99.80
L	0.05	0.18	99.76
M	0.08	0.50	99.43
N	0.05	0.42	99.52
O	0.05	0.64	99.31
P	0.04	0.48	99.47
Q	0.15	0.58	99.27
R ^d	--	--	--

^aDerived from mass spectrometric analysis of gas collected from rod plenum.

^bDerived from in-line radiation monitor data from shear cell exhaust stack.

^cDerived from mass spectrometric analysis of dissolver off-gas samples.

^dFission-gas distribution could not be calculated for Rod R since dissolver off-gas was not sampled as a result of pump problems (see Section IV.E.3.c); only minor traces of fission gas were released during plenum puncture and shearing of Rod R.

preparing for execution of the EOL campaign. Careful forethought in planning the EOL operations and in the design of equipment and procedures, together with extensive development and characterization of the operating systems once they were in place, provided a high level of confidence that these project goals would be achieved.

Execution of the EOL campaign was not free of problems, but the problems that arose were remarkably few considering the difficulties associated with shearing 17 irradiated fuel rods into segments, dissolving 152 fuel-rod segments, and performing several thousand analyses on the dissolver solutions and gas samples in a hot-cell environment over a 15-month period.

Our review and compilation of EOL data for preparing the error analyses presented in this final report have led us to conclude that the limits to probable

Fig. 31. Correlation Between Fission Gas and ^{137}Cs in Rod Segments

error we have quoted for each reported result are properly derived and correctly calculated. On the basis of this conclusion, we are confident that all of the results we provided are valid and of a quality that satisfies the needs of the LWBR-POB parent program.

I. Patents

A list of the patents resulting from this work is given below:

1. Can-Out Hatch Assembly and Positioning System
P. J. Basner, R. C. Frank, and J. C. Hoh
Statutory Investigation Registration No. H11, issued
January 7, 1986
2. Can-Out Hatch Assembly with Magnetic Retention Means
R. C. Frank and J. C. Hoh
Statutory Investigation Registration No. H10, issued
January 7, 1986
3. Material Bagging Device
C. G. Wach, R. E. Nelson, and S. B. Brak
Patent No. 4,480,426, issued November 6, 1984
4. Accordion-Folded Boot Shield for Flexible Swivel Connection
J. C. Hoh
Patent No. 4,607,852, issued August 26, 1986

5. Bag-Out Material Handling System
S. B. Brak and H. F. Milek
Patent No. 4,489,538, issued December 25, 1984
6. Bag-Out Material Handling System
S. B. Brak
Patent No. 4,493,179, issued January 15, 1985

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The scope and duration of a project such as this, which dates to 1976, make recognition of all the participants problematic. We wish, therefore, to also acknowledge the efforts of the many unnamed persons who played a vital role in the overall technical success of this work.

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APPENDIX A

CALCULATION OF CUT PLAN FOR SHEARING A FUEL ROD SEGMENT

The objective of the cut-plan calculation is to identify the number of cuts at each of two cut lengths, which differ by 0.001 in., that will exactly span the distance between the as-cut boundary of the previous segment and the desired boundary of the segment to be sheared, and yet keep each cut length close to the nominal value specified for the segment in the fuel rod information file (FRIF). To accomplish this objective, the OPRFSS operating program carried out the following computations.

First, the distance between the segment boundaries (i.e., the desired length of the segment, SL) was computed as the difference between the desired segment boundary as read from the FRIF and the actual boundary location that was measured when the previous segment was sheared. If the segment was the first to be cut from the rod, the previous boundary was taken as the end of the rod. Next, the total number of cuts, TNC, to be made in shearing the segment was calculated using the relationship

$$TNC = \text{INT}(SL/NCL + 0.999999) \quad (\text{A-1})$$

where NCL is the nominal cut length specified in the FRIF, and the mathematical operator INT indicates that the term in parentheses is to be truncated to the next lower integer. The near-unity constant on the right-hand side of Eq. A-1 ensures that if the segment length is close to an exact multiple of the nominal cut length, the calculated cut length will correspond to the nominal value.

Having determined the total number of cuts to be made in shearing the segment, the program calculated the length, CL1, to be used for the shorter cuts as follows:

$$CL1 = \text{INT}(SL/TNC) \quad (\text{A-2})$$

The length, CL2, of the longer cuts was calculated from

$$CL2 = CL1 + 0.001 \quad (\text{A-3})$$

The number of cuts to be made at each length, N1 and N2, respectively, for CL1 and CL2, was then computed as follows:

$$N2 = \frac{TNC(SL/TNC - CL1)}{0.001} \quad (\text{A-4})$$

and

$$N1 = TNC - N2 \quad (A-5)$$

In principle, making N1 cuts at length CL1 and N2 cuts at length CL2 should provide a segment with length equal to SL within 0.001 in. In practice, because no adjustments were made to the calculated cut lengths for thermal expansion of the shear base, the length of the last cut (the segment boundary cut) had to be adjusted slightly to compensate for temperature-related variations in the length equivalent of the absolute shaft transducer (AST) reading. This adjustment was made automatically by the computer just prior to making the boundary cut so that the machine base temperature (measured by the computer and used in the calculation) would be appropriate for the time when the cut was actually made. The magnitude of the change in cut length that resulted from this compensation for thermal expansion was typically quite small; for example, for a 25-in. segment (the longest segment sheared from any LWBR fuel rod), thermal expansion of the shear machine base typically required an adjustment of the length of the final cut of only 0.003 in. Thus, the length of the last cut did not differ substantially from the nominal cut length, even in the worst-case situation.

APPENDIX B

ESTIMATION OF BIAS AND STANDARD DEVIATION
FOR URANIUM ISOTOPE ABUNDANCESA. Introduction

The weight percent abundances of the uranium isotopes in each dissolved segment were determined by analysis of the uranium in solution from the unspiked blend tank (UBT). For each segment, single aliquots were taken from each of two separate samples of the UBT solution, and ratios of each uranium isotope to ^{233}U were measured for each aliquot. For ^{232}U , these ratios were measured by alpha pulse height analysis (APHA) as described in Section IV.F.1.d.(2) of the report body; for the other isotopes, the ratios were measured by thermal ionization mass spectrometry (TIMS) as described in Section IV.F.1.d.(1). In each case, the averages of the two corresponding ratios measured for a given segment were used in computing the weight percent abundances for that segment according to

$$\text{WPC}_{233} = \frac{100\%}{1 + \sum R_j}, j = 232, 234, 235, 236, 238 \quad (\text{B-1a})$$

$$\text{WPC}_i = \frac{R_i (100\%)}{1 + \sum R_j}, i, j = 232, 234, 235, 236, 238 \quad (\text{B-1b})$$

Here, WPC_i stands for the weight percent abundance of isotope i ; R_j stands for the measured weight ratio $^i\text{U}/^{233}\text{U}$; and $\sum R_j$ is the sum of measured ratios for all isotopes (other than ^{233}U) present in the segment uranium.

The weight percent abundances that ANL reported to BAPL during the EOL campaign were calculated using a computer program that not only compiled the APHA and TIMS ratio data required for the calculations, but also carried out a statistical analysis of the ratio data and propagated the resulting error estimates to provide an estimate of the uncertainty associated with each reported isotope abundance. The uncertainty quoted for each result represented an estimate of the standard deviation of the reported value and included contributions from "randomly varying systematic errors" (i.e., errors that affected all measurements on a given isotope in the segment to about the same extent, but may have varied randomly between segments) as well as from random errors that could be evaluated from observed differences between the duplicate measurements performed for each isotope of each segment.

In this appendix, we explain how the uncertainties quoted for the individual uranium isotopic results were derived and estimate the potential bias associated with each reported result. Because the weight percent abundance of each isotope in each segment was not measured directly but was calculated from measured ratio data, we begin with a discussion of errors associated with the ratio measurements and then propagate these errors to the calculated parameters in a conventional way.

Uncertainties in measured isotope ratios arise primarily from two sources: (1) uncertainties in the actual measurements and (2) uncertainties associated with any differences that might exist between the isotopic composition of the sample on which the measurements are made and the segment uranium whose composition is desired. Such differences usually result from contamination of the sample, during its preparation and handling, with traces of uranium having a widely different isotopic composition from that of the sample. The ANL procedures for the EOL work were carefully designed to minimize the potential for such contamination and to mitigate this contamination if it occurred at expected trace levels. For example, whenever practical, the size of the UBT solution aliquot prepared for use in the isotope ratio measurements was chosen to contain a relatively large amount (1 mg) of segment uranium so as to "swamp out" the effects of trace isotopic contaminants. In addition, all-new glassware, ion-exchange columns, etc., were used in processing each sample.

This is not to say that all contamination effects were, or could have been, eliminated. Of particular importance in this regard is the effect of introducing trace quantities of highly enriched uranium from mass spectrometric isotopic dilution (MSID) spikes, or from other sources, into the isotopic samples from low-uranium segments where an aliquot large enough to contain 1 mg U could not be practically obtained or handled. Such contamination may be referred to as an isotopic procedure blank.

Errors associated with both the isotope ratio measurements and the isotopic procedure blank have systematic as well as random components. Errors from the mass spectrometric measurement, the isotopic procedure blank, and the APHA measurement are considered separately in the following section.

B. Errors in Measured Uranium Isotope Ratios

1. Errors in Mass Spectrometric Ratio Measurements

In TIMS, each measured ratio of ion intensities must be corrected for an inherent bias caused by isotopic fractionation that accompanies evaporation of the sample during analysis. The magnitude of the correction is determined by a comparison of measured and certified ratios for a series of NBS isotopic standard reference materials (SRMs). In the EOL campaign, the fractionation correction was determined from a running average of ten analyses of NBS SRMs chosen from the series U-010 through U-930; NBS U-500 was run as one of these standards at least once for every five segments as called for in the project requirements. Each of these isotopic SRMs is a mixture of ^{235}U and ^{238}U in a certified ratio. For U-500, which has an atom ratio of ^{235}U to ^{238}U of 0.9997, the ratio is known to at least 0.1%, relative, according to the NBS certificate. On a one-sigma basis the uncertainty in this standard ratio can be taken as $\pm 0.05\%$.

Evaluation of the fractionation correction that must be applied in the case of a specific measured isotope ratio is complicated by the fact that the magnitude of the appropriate correction depends upon the masses of the nuclides in the ratio, whereas NBS SRMs are available only with ^{235}U and ^{238}U as major isotopes suitable for sensitive measurement of the fractionation effect. For uranium isotopes, the magnitude of this effect can be taken as proportional to the

difference between the mass numbers of the isotopes in a given ratio. For a given pair of isotopes ^iU and ^jU , the appropriate correction factor for each ratio is given by

$$C_{i/j} = 1 + \left(\frac{i - j}{3} \right) E_0 \quad (\text{B-2})$$

where E_0 is evaluated from the NBS SRMs containing ^{235}U and ^{238}U :

$$\frac{\text{Certified } ^{235}\text{U}/^{238}\text{U}}{\text{Measured } ^{235}\text{U}/^{238}\text{U}} = \frac{R_{\text{cert.}}}{R_{\text{meas.}}} = 1 + E_0 \quad (\text{B-3})$$

The corrected ratio is obtained by multiplying the measured ratio by the appropriate $C_{i/j}$:

$$R_{\text{corr, } i/j} = R_{\text{meas, } i/j} C_{i/j} \quad (\text{B-4})$$

Fractionation corrections applied to the measured isotope ratios eliminate the largest part of the bias error inherent in the ratio measurement. Uncertainty in the corrected ratio remains, however, from three sources: (1) uncertainties in the certified value of the standards used in determining the magnitude of the correction factor; (2) uncertainties in the measurements made in establishing the magnitude of the correction (i.e., in measuring $R_{\text{meas.}}$ in Eq. B-3); and (3) uncertainties in measuring the (unknown) sample ratio, $R_{\text{meas, } i/j}$, in Eq. B-4.

Uncertainty in the certified value of the standards represents a potential error in the value determined for the correction factor and propagates to a potential bias in the corrected ratio. The magnitude of this bias depends on the identity of the nuclides involved in a specific ratio, as may be deduced from the foregoing treatment. Potential bias errors in the corrected ratios are evaluated in Section a, below.

Uncertainty in the measurements used to establish the correction factors for ratios of individual isotopes arises from random errors associated with measurements of the apparent ratios for the isotopic SRMs. These errors propagate to a variance in the corrected ratio for a sample. This variance may represent a systematic error in the corrected ratios for samples processed over a short term, since the same corrections would apply to all replicates and the error would have the same magnitude and direction. In the long term, however, this error would vary randomly from time to time and is appropriately treated as a contributor to the standard deviation of the corrected ratio. Its magnitude cannot be readily estimated from replicate data obtained from analysis of the samples but must be estimated by other means. Our approach to estimating this variance term is described in Section b, below.

Uncertainties associated with the measured (i.e., uncorrected) ratios for the samples were treated as purely random errors. As such, their contributions

to the variance of the corrected ratios are included in the standard deviation that was estimated by the analysis of variance of replicate data on the segment uranium samples for each LWBR fuel rod. This analysis was performed by the report-generating computer program and is described at the end of Section B.2, below.

a. Bias in the Uranium Isotope Ratios Measured by Mass Spectrometry

To the extent that the certified value of the standard ratio of each isotopic SRM is not exactly equal to the true value, a small residual bias in the mass spectrometrically determined ratios may exist even after corrections based on the SRMs are made. A limit to the magnitude of this residual bias may be estimated from the uncertainty associated with the standards used in evaluating the correction factor. If $R_{\text{cert.}}$ has a one-sigma uncertainty of $\pm 0.05\%$, as for SRM U-500, then the possible bias in the corrected ratio, $R_{i/j}$, is

$$B_R(R_{i/j}) = \left(\frac{i - j}{3} \right) (\pm 0.05\%) \quad (\text{B-5})$$

where $B_R(Y)$ represents the relative bias in the parameter Y .

For the individual ratios, R_i , of isotope ^iU to ^{233}U as determined during the EOL campaign, Eq. B-5 gives the following limits to potential bias in the measured ratios:

$$\begin{aligned} B_R(R_{234}) &= \frac{1}{3} (\pm 0.05\%) = 0.017\% \\ B_R(R_{235}) &= \frac{2}{3} (\pm 0.05\%) = 0.033\% \\ B_R(R_{236}) &= \frac{3}{3} (\pm 0.05\%) = 0.05\% \\ B_R(R_{238}) &= \frac{5}{3} (\pm 0.05\%) = 0.083\% \end{aligned} \quad (\text{B-6})$$

For these estimated limits to potential bias to be valid, one must assume that the corrections applied to the EOL measurements actually did eliminate bias relative to the isotopic SRMs and, in particular, relative to U-500. That this assumption is justified may be demonstrated by consideration of data obtained for measurements on the U-500 standard. In Appendix C, we list results obtained by treating samples of SRM U-500 as unknowns in the TIMS measurement procedure. Each listed value was calculated from a measured ratio that was corrected using the factor derived from the previous ten standard analyses, just as an unknown ratio would be calculated. The data in

Appendix C include 96 analyses of the U-500 SRM performed over the time period from December 1982 through October 1985. The average of these 96 runs differs from the certified ratio for this SRM by only 0.005%, which demonstrates that residual bias relative to U-500 is indeed negligible. The standard deviation of an individual measurement based on these results is 0.043%, which attests to the long-term stability of the ANL ratio measurements. From these observations, it may be confidently concluded that bias in the mass spectrometric uranium isotope ratios from sources other than uncertainty in the standard is truly negligible.

b. Estimation of Systematic Variance from Correction of Measured Ratios

By substituting Eq. B-2 into Eq. B-4, one obtains the following expression for the corrected ratio in terms of the measured variables, $R_{\text{meas}, i/j}$ and E_0 :

$$R_{\text{corr}, i/j} = R_{\text{meas}, i/j} + R_{\text{meas}, i/j} \left(\frac{i - j}{3} \right) E_0 \quad (\text{B-7})$$

Taylor-series expansion of this expression then provides the approximate relationship to describe the variance, $\sigma^2(R_{\text{corr}, i/j})$, of the corrected ratio:

$$\sigma^2(R_{\text{corr}, i/j}) = \left[1 + \left(\frac{i - j}{3} \right) E_0 \right]^2 \sigma^2(R_{\text{meas}, i/j}) + \left[\left(\frac{i - j}{3} \right) R_{\text{meas}, i/j} \right]^2 \sigma^2(E_0) \quad (\text{B-8})$$

Here, and in the remainder of this appendix, the notation $\sigma^2(X)$ stands for the absolute variance of the variable X ; $\sigma_R^2(X)$ stands for the relative variance.

The first term on the right-hand side in Eq. B-8 corresponds to the random component of the variance. The second term may be taken as the systematic component since, in the short term, the value of E_0 applied to a series of measurements is a constant determined from the average of ten prior standard analyses, and its variance (equal to zero) will not contribute to the observed variance of the corrected ratios. In the long term, however, the variance of E_0 may be obtained from the variance of the mean of the correction factors obtained from the ten standard analyses that were used in its evaluation.

For the purpose of estimating the systematic variance from correction of the measured ratios in the computer program that estimated uncertainties in the EOL results, the percent relative standard deviation (%RSD) for a single ratio measured from analysis of an NBS SRM was taken to be a constant, 0.042%, evaluated from a statistical analysis of 72 separate standard runs performed prior to writing the program. That this estimated standard deviation continued to be applicable during the EOL campaign may be deduced from the data in Appendix C, where the %RSD of 96 runs on NBS U-500, including runs performed in the campaign, is found to be 0.043%. We note that, although these data are presented in the form of corrected ratios, the RSD estimated from them provides a good approximation of the standard deviation of the measured ratios because the systematic variance contribution is small, as shown below.

The %RSD of the mean of ten measurements on the isotopic SRMs, each having a %RSD of 0.042%, is given by $0.042\%/\sqrt{10} = 0.0133\%$. This %RSD corresponds to the %RSD of the $1 + E_0$ term in Eq. B-3, and thus represents a standard deviation of the parameter E_0 of 0.000133. Given this value and the functional form of the systematic-error term in Eq. B-8, systematic variances in the mass spectrometrically measured isotope ratios, designated $\sigma_{\text{SYS,MS}}^2$, were assigned as follows:

$$\begin{aligned}\sigma_{\text{SYS, MS}}^2 (R_{234}) &= (0.0000443)^2 R_{234}^2 \\ \sigma_{\text{SYS, MS}}^2 (R_{235}) &= (0.0000886)^2 R_{235}^2 \\ \sigma_{\text{SYS, MS}}^2 (R_{236}) &= (0.000133)^2 R_{236}^2 \\ \sigma_{\text{SYS, MS}}^2 (R_{238}) &= (0.000222)^2 R_{238}^2\end{aligned}\tag{B-9}$$

The report-generating computer program evaluated this variance contribution for each measured ratio and combined it with other appropriate variances to provide an estimate of the total variance in each ratio. These other variances included a systematic variance associated with the isotopic procedure blank and a random variance that was evaluated from replicate data obtained during processing of segments from each fuel rod. Estimation of these other variances is described in the following sections.

2. Errors from Isotopic Procedure Blanks

The uranium samples used in determining the measured uranium isotope ratios actually represent a mixture of uranium from the dissolved fuel rod segment and uranium arising from the isotopic procedure blank. The ratio, $R_{\text{mix, i/j}}$, of isotopes ${}^i\text{U}/{}^j\text{U}$ for this mixture may be described by the relationship

$$R_{\text{mix, i/j}} = \frac{{}^i\text{U}_{\text{seg}} + {}^i\text{U}_{\text{Blank}}}{{}^j\text{U}_{\text{seg}} + {}^j\text{U}_{\text{Blank}}}\tag{B-10}$$

Here, the variable U represents the weight of the superscripted uranium isotope from the subscripted source.

Rearranging Eq. B-10 and substituting variables give the expression

$$R_{\text{mix, i/j}} = R_{\text{seg, i/j}} + \frac{{}^i\text{U}_{\text{Blank}}}{{}^j\text{U}_{\text{seg}}} - R_{\text{mix, i/j}} \left(\frac{{}^j\text{U}_{\text{Blank}}}{{}^j\text{U}_{\text{seg}}} \right)\tag{B-11}$$

Here, $R_{\text{seg, i/j}}$ (equal to ${}^i\text{U}_{\text{seg}}/{}^j\text{U}_{\text{seg}}$) is the sought-for ratio of isotopes in the segment. Since no corrections for the effects of the isotopic blank were incorporated into the ANL computational procedures, the difference between $R_{\text{mix, i/j}}$ and $R_{\text{seg, i/j}}$, obtainable from Eq. B-11, represents a potential bias error in the reported segment ratios if the blank levels of ${}^i\text{U}$ and ${}^j\text{U}$ are significant. To account for this error, a variance term was added to the estimated variance

of each measured isotope ratio. This additional variance was designated the systematic variance due to the isotopic blank, $\sigma_{\text{SYS, Blank}}^2$, and was calculated by the report-generating computer program based on the following assumptions and approximations. First, it was assumed that uranium in the procedure blank contained only the isotopes ^{233}U , ^{235}U , and ^{238}U , and further, that the average level of contamination by each of these isotopes was close to that of the others. This assumption was based on a consideration of the likely sources of uranium in the blank, given the expectation that significant levels of contamination should arise only from the introduction of materials handled routinely in preparing the samples. The primary uranium-bearing materials in this category are segment uranium ($>80\%$ ^{233}U), NBS 950a spike ($>99\%$ ^{238}U), and NBS 993 spike ($>99\%$ ^{235}U). Thus, we applied the approximations that

$$\begin{aligned} {}^{232}\text{U}_{\text{Blank}} &= {}^{234}\text{U}_{\text{Blank}} = {}^{236}\text{U}_{\text{Blank}} = 0 \\ {}^{233}\text{U}_{\text{Blank}} &= {}^{235}\text{U}_{\text{Blank}} = {}^{238}\text{U}_{\text{Blank}} \end{aligned} \quad (\text{B-12})$$

The magnitude of the procedure blank for ^{233}U , ^{235}U , and ^{238}U was estimated from data obtained for a number of samples analyzed prior to the EOL campaign, including samples that were prepared by processing unadulterated MSID spikes and samples for which only blank levels of uranium were expected. These latter samples were analyzed during processing of the GRIP-II Validation Rod (No. 79-440). The resulting estimated average blank was 6×10^{-8} g uranium.

To put this value in perspective, we note that a blank of this size in an aliquot containing 1 mg of ^{233}U would increase the ^{235}U or ^{238}U isotopic abundance by 0.006%, absolute; if the aliquot contained only 0.1 mg ^{233}U , the resulting increase in these abundances would be 10 times larger, or 0.06%. Thus, even though the blank levels are small, their influence on the isotopic measurements can be substantial. Post-EOL examination of data from low-level uranium samples such as plenum analyses, BT rinse samples, etc., suggested that 6×10^{-8} g uranium may actually overestimate the amounts of ^{233}U and ^{238}U in the isotopic procedure blanks pertinent to the EOL campaign. Results for ^{235}U from isotopic measurements on plenum-segment uranium samples were consistent with the pre-EOL estimate.

The systematic variance due to the isotopic procedure blank was computed in the report-generating program for each measured ratio, using an expression derived by a Taylor-series expansion of Eq. B-11 that approximates the relationship between the variance of the difference between $R_{\text{mix}, i/j}$ and $R_{\text{seg}, i/j}$ and the blank levels of the isotopes in the ratio. This relationship is

$$\sigma_{\text{SYS, Blank}}^2(R_{i/j}) = \frac{{}^i\text{U}_{\text{Blank}}^2}{{}^j\text{U}_{\text{seg}}^2} + \frac{\left(R_{\text{mix}, i/j} {}^j\text{U}_{\text{Blank}}\right)^2}{{}^j\text{U}_{\text{seg}}^2} \quad (\text{B-13})$$

For this relationship, ${}^j\text{U}_{\text{Blank}}$ (equal to ${}^{233}\text{U}_{\text{Blank}}$ in all cases) was assigned the value 6×10^{-8} g, in keeping with the above discussion. Similarly, ${}^i\text{U}_{\text{Blank}}$ was given the same value when $i = 235$ or 238 but was set to zero for other isotopes. The value of ${}^{233}\text{U}_{\text{seg}}$ was calculated in the computer program as the root mean square (RMS) weight of this isotope in the aliquots taken for each isotopic analysis of a particular segment's uranium; the weight of ^{233}U in each

individual aliquot was calculated from (1) the concentration of U in the corresponding UBT solution, as determined in the segment assay procedures, (2) the weight fraction of ^{233}U in this uranium, as measured for the segment of interest, and (3) the weight of the aliquot taken for analysis.

For most EOL samples, in particular those for which 1 mg of segment uranium was present in the aliquot, $\sigma_{\text{SYS, Blank}}$ made only a small contribution to the estimated uncertainty of the measured ratios. For samples where smaller amounts of uranium were present in the aliquots that were analyzed, this error contribution increased in importance, becoming the primary source of uncertainty when the aliquot contained only 100 μg U or less.

Besides the potential systematic error associated with the isotopic procedure blank, we also considered the random errors that arise from variations in the blank levels for each individual isotope. These variations contribute to the observed variance in the measured ratios through a variance term that has a similar functional form to that of Eq. B-13. Thus, the random variance of the measured ratios was presumed to follow an expression of the form:

$$\sigma_{\text{OBS}}^2 = \sigma_{\text{NORM}}^2 + \left(\frac{1}{^{233}\text{U}_{\text{ALQ}}^2} \right) \sigma_{\text{LOW U}}^2 \quad (\text{B-14})$$

In this expression, σ_{OBS}^2 represents the apparent variance of a set of replicate measurements for which the isotope ratios are identical and the weight of ^{233}U in the aliquots taken for analysis is the same. We substituted the variable designation $^{233}\text{U}_{\text{ALQ}}$ for the designation $^{233}\text{U}_{\text{seg}}$, as used in Eq. B-13 for this weight of ^{233}U , to emphasize that it stands for the weight of ^{233}U in the aliquot and not that in the entire segment. The variance σ_{NORM}^2 includes all random variance contributions to the observed variance except those that are inversely proportional to $^{233}\text{U}_{\text{ALQ}}^2$; these contributions include variances from isotopic inhomogeneity of the UBT solution, mass spectrometer measurement errors, and any other random error source, whether identified or not. The final variance term in Eq. B-14 accounts for the increase in uncertainty of the measured isotope ratio when the weight of ^{233}U in the aliquot becomes small; $\sigma_{\text{LOW U}}^2$ may be interpreted as a measure of variations in the blank levels of the isotopes of interest among the samples in a set of replicates.

The individual variances, σ_{NORM}^2 and $\sigma_{\text{LOW U}}^2$, were estimated in the computer program by pooling replicate data on the ratios measured for each segment of an individual fuel rod and then using the following algorithms. First, the apparent variance of replicate (usually duplicate) ratio measurements performed for each isotope of each segment was computed. Next, the individual variances for a particular isotope were summed, weighting the variance from each segment according to the square of the RMS weight of ^{233}U present in the aliquots used in making measurements for a given segment. Based on Eq. B-14, this sum may be related to σ_{NORM}^2 and $\sigma_{\text{LOW U}}^2$ as follows:

$$\sum_{\text{seg}} (^{233}\text{U}_{\text{ALQ, seg}}^2 \sigma_{\text{OBS, seg}}^2) = \left(\sum_{\text{seg}} ^{233}\text{U}_{\text{ALQ, seg}}^2 \right) \sigma_{\text{NORM}}^2 + N_{\text{seg}} \sigma_{\text{LOW U}}^2 \quad (\text{B-15})$$

Here, N_{seg} is the number of segments for which isotope ratio data are available in the rod under consideration. Because, for our work, the second term on the right side of this expression was expected to be small relative to the first term, it was set to zero and an approximate value for σ_{NORM}^2 was computed:

$$\sigma_{\text{NORM}}^2 = \frac{\sum_{\text{seg}} ({}^{233}\text{U}_{\text{ALQ, seg}}^2 \sigma_{\text{OBS, seg}}^2)}{\sum_{\text{seg}} {}^{233}\text{U}_{\text{ALQ, seg}}^2} \quad (\text{B-16})$$

In similar fashion, the observed variances for each segment were summed again, this time without weighting the values for the individual segments. Again, if Eq. B-14 is used as a model for interpretation, this sum can also be related to σ_{NORM}^2 and $\sigma_{\text{LOW U}}^2$ as follows:

$$\sum_{\text{seg}} \sigma_{\text{OBS, seg}}^2 = N_{\text{seg}} \sigma_{\text{NORM}}^2 + \left(\sum_{\text{seg}} \frac{1}{{}^{233}\text{U}_{\text{ALQ, seg}}^2} \right) \sigma_{\text{LOW U}}^2 \quad (\text{B-17})$$

Since σ_{NORM}^2 was known from application of Eq. B-16, this expression could be solved for $\sigma_{\text{LOW U}}^2$:

$$\sigma_{\text{LOW U}}^2 = \frac{(\sum_{\text{seg}} \sigma_{\text{OBS, seg}}^2) - (N_{\text{seg}} \sigma_{\text{NORM}}^2)}{\left(\sum_{\text{seg}} \frac{1}{{}^{233}\text{U}_{\text{ALQ, seg}}^2} \right)} \quad (\text{B-18})$$

Values for σ_{NORM} and $\sigma_{\text{LOW U}}$ obtained in this way for each mass spectrometrically measured isotope ratio in each LWBR fuel rod are shown in Tables B-1 and B-2, respectively.

As Table B-1 shows, the magnitude of σ_{NORM}^2 was generally quite small, corresponding to a standard deviation of only a few parts per million, absolute, in the ratios measured for the minor isotopes ${}^{236}\text{U}$ and ${}^{238}\text{U}$. Somewhat larger standard deviations applied for the major isotopes, ${}^{234}\text{U}$ and ${}^{235}\text{U}$, but on a relative basis these correspond to less than 0.1% of the measured ratio in all cases. These error levels are consistent with those expected from the mass spectrometric measurements that were made in determining the ratios: signal noise in the TIMS measuring systems introduces a random error of a few parts per million in all ratio measurements, while a proportional error of a few hundredths of a percent arises from fractionation differences among samples (see discussion earlier in this appendix).

Values for $\sigma_{\text{LOW U}}$, as shown in Table B-2, are similarly small, with the largest values (on the order of 10^{-8}) found for ratios involving ${}^{235}\text{U}$ and ${}^{238}\text{U}$. These values are consistent with our expectation (see earlier discussion) that significant contamination of the sample aliquots by these isotopes would be most likely.

Table B-1. Values of the Random Standard Deviation σ_{NORM} Estimated from Isotope Ratios Measured for LWBR Fuel Rods

Rod Designation	N_{seg}^a	$\sigma_{\text{NORM}} \times 10^6$			
		234/233	235/233	236/233	238/233
B	7	8.5	6.1	1.6	7.0
C	7	15.6	9.0	3.2	3.1
D	7	9.2	54.0	3.6	7.4
E	7	4.3	16.0	1.9	6.7
F	8	4.5	12.7	0.7	3.4
G	7	7.4	10.6	1.6	8.6
H	15	8.4	25.4	2.1	5.6
I	8	30.1	16.6	2.5	4.2
J	8	9.1	51.7	1.7	3.8
K	8	9.1	13.6	1.9	2.4
L	8	13.9	31.6	1.7	4.9
M	6	30.2	8.0	1.7	11.8
N	6	12.0	19.3	1.9	10.6
O	6	13.7	9.4	1.5	6.4
P	6	21.4	9.8	2.4	14.5
Q	6	12.7	24.5	1.3	4.5
R	15	3.6	20.2	1.4	3.1

^aNumber of segments for which replicate isotope ratio data were available.

3. Estimation of Total Variance of Isotope Ratios Measured by Mass Spectrometry

The total variance of each mass spectrometrically measured isotope ratio for each fuel rod segment was estimated in the report-generator program by combining variance terms associated with each of the error sources that were introduced in the foregoing discussion. Thus, the total variance, for each ratio, $\sigma_{\text{TOT}}^2(R_{i, \text{seg}})$, was calculated as

$$\sigma_{\text{TOT}}^2(R_{i, \text{seg}}) = \sigma_{\text{SYS, MS}}^2(R_i) + \sigma_{\text{SYS, Blank}}^2(R_{i, \text{seg}}) + \left[\sigma_{\text{NORM}}^2 + \left(\frac{1}{^{233}\text{U}_{\text{ALQ, seg}}^2} \right) \sigma_{\text{Low U}}^2 \right] / N_{\text{ISO}} \quad (\text{B-19})$$

where N_{ISO} represents the number of independent samples of the UBT solution that were analyzed in obtaining the average ratio used as the appropriate value of $R_{i, \text{seg}}$ in subsequent calculations. Dividing the random variance, represented by the term in brackets, by N_{ISO} accounts for the reduction in variance that accompanies the use of this average value. The computer program made a separate estimate of variance for each measured isotope ratio, taking into account the dependence of this variance on the actual value of the ratio, the size of the aliquot taken in preparing each sample for analysis, etc. A discussion of individual results of this error treatment would, therefore, be overwhelming.

Table B-2. Values of the Random Standard Deviation $\sigma_{\text{LOW U}}$ Estimated from Isotope Ratios Measured for LWBR Fuel Rods

Rod Designation	N_{seg}^a	$\sigma_{\text{LOW U}} \times 10^8$			
		234/233	235/233	236/233	238/233
B	7	0.00	0.00	0.08	0.36
C	7	0.00	2.06	0.00	0.12
D	7	0.00	0.00	0.00	0.34
E	7	0.00	0.00	0.00	0.00
F	8	0.00	2.38	0.02	0.63
G	7	0.00	1.01	0.00	1.86
H	15	0.00	3.82	0.05	0.59
I	8	0.00	0.00	0.00	0.00
J	8	0.60	4.46	0.05	0.32
K	8	0.00	1.64	0.00	0.00
L	8	0.00	0.00	0.00	0.28
M	6	0.54	0.16	0.10	0.25
N	6	0.00	0.00	0.00	0.00
O	6	0.36	0.00	0.03	0.25
P	6	0.08	0.52	0.05	0.24
Q	6	0.23	0.88	0.01	0.15
R	15	0.00	0.97	0.01	0.10

^aNumber of segments for which replicate isotope ratio data were available.

Having described the way in which the individual error terms were derived and calculated and having presented information to support the validity of assumptions involved in these derivations, we believe it sufficient to say that the variances estimated through the programmed calculations represent a complete and realistic description of non-bias errors associated with the isotope ratios measured by mass spectrometry.

4. Estimation of Errors in Alpha Pulse Height Measurement of $^{232}\text{U}/^{233}\text{U}$ Weight Ratio

In ANL's APHA procedure for measuring the ratio of ^{232}U to ^{233}U in a sample of the uranium from a given segment, the ratio of α -counts having a peak energy of 5.3 MeV to those having a peak energy at 4.8 MeV was measured. The peak at 5.3 MeV represents counts obtained from the alpha decay of ^{232}U , while that at 4.8 MeV contains counts from decay of both ^{233}U and ^{234}U . Because the efficiency of the alpha detector is constant over the energy range over which these peaks extend, this count ratio is identical to the ratio of disintegration rates of the nuclides that contribute to each peak.

The count ratio, R_{count} , is converted to the weight ratio $^{232}\text{U}/^{233}\text{U}$ by using the mass spectrometrically measured $^{234}\text{U}/^{233}\text{U}$ weight ratio and the relationship

$$R_{\text{count}} = \frac{W_{232} SA_{232}}{W_{233} SA_{233} + W_{234} SA_{234}} \quad (\text{B-20})$$

where W_i is the weight of ^iU in the sample on the planchet examined by APHA, and SA_i is the specific activity of isotope ^iU . Values for SA_i in Eq. B-20 were derived from the half-lives of ^{232}U (71.8 yr), ^{233}U (1.589×10^5 yr), and ^{234}U (2.459×10^5 yr).

The actual formula used in calculating the $^{232}\text{U}/^{233}\text{U}$ weight ratio was obtained by rearranging Eq. B-20:

$$R_{232} = \frac{R_{\text{count}} [1 + (SA_{234}/SA_{233}) R_{234}]}{SA_{232}/SA_{233}} \quad (\text{B-21})$$

Errors in R_{232} arise from errors in the count ratio, R_{count} , the mass spectrometric weight ratio R_{234} , and the half-life values used to obtain SA_i . The count ratio was taken as contributing only random errors to the error in R_{232} since no mechanism for introducing a systematic error in R_{count} exists if the individual alpha peaks are well separated, as they were for our EOL work. This random error in R_{count} was estimated on the basis of counting statistics that described the uncertainty in the number of counts accumulated in each peak taken for the ratio and was automatically computed by the computer program that processed the APHA spectrum obtained for each sample. In general, this counting error amounted to a few tenths of a percent, RSD.

Error in the mass spectrometric ratio, R_{234} in Eq. B-21, was shown to be negligibly small relative to errors from other sources and was not included in estimates of errors associated with R_{232} .

Errors in the half-life values used in calculating the SA_i give rise to systematic (i.e., bias) error in R_{232} . Standard mixtures of ^{232}U and ^{233}U are not available for use in evaluating such systematic errors. However, for some segments from GRIP II Rod 79-440, which was processed in pre-EOL demonstration work, the thorium content was sufficiently low to allow confident measurement of the $^{232}\text{U}/^{233}\text{U}$ ratio by mass spectrometry. A comparison of the results from the mass spectrometric and APHA measurements on uranium samples from these segments suggested that a bias of 3.1% (the mass spectrometric value being lower) existed between the methods. Because of the paucity of data from these measurements, we did not apply any bias corrections to the EOL APHA data but included a systematic variance term, corresponding to a standard deviation of 3.1%, in our estimate of the standard deviation of each measured R_{232} .

Late in the EOL campaign we discovered that the value we had used for the half-life of ^{232}U may not have been correct. Recent listings of the half-lives of uranium isotopes recommend a value for ^{232}U of 69 ± 1 yr based on

recent measurements by Aggarwal et al.* Our EOL data indirectly support this value for the ^{232}U half-life, as shown in the following discussion.

Although we did not use mass spectrometer values for the $^{232}\text{U}/^{233}\text{U}$ ratios in computing our EOL results, these were routinely measured for each sample analyzed. We anticipated that the TMS ratio, R_{ms} , for each sample would be equal to or greater than the corresponding APHA ratio, R_{a} , since ^{232}Th in the samples would increase the observed TMS ratio. However, the TMS ratio proved to be smaller than the APHA ratio in a large majority of cases, particularly for samples having a relatively high ^{232}U content, as illustrated in Fig. B-1a. In Fig. B-1b, we have plotted our data using a value of 69.5 yr for the ^{232}U half-life. The absence of significant negative differences ($R_{\text{ms}} - R_{\text{a}}$) in this plot demonstrates the consistency of this half-life with our results. These data strongly indicate that the $^{232}\text{U}/^{233}\text{U}$ ratios measured by ANL during the EOL campaign are subject to a bias on the order of +4%, relative. Because the ^{232}U content of the EOL fuel rods was small (<0.3% by weight of the total uranium) and because our measurement of the ^{232}U isotopic abundance was not subject to specific requirements, this bias error is a matter of curiosity only and does not prohibit ANL's meeting any error requirements. We do, however, include this bias among the contributions to potential bias error in the weight percent abundance values estimated by the method described below.

Variance in the measured R_{232} values was estimated by the report-generator computer program by summing variance terms from several sources, in a manner similar to that used for the mass spectrometric ratios (cf. Eq. B-19). The expression describing this variance is

$$\sigma_{\text{TOT}}^2 (R_{232, \text{seg}}) = \sigma_{\text{SYS, Blank}}^2 (R_{232}) + \sigma_{\text{count}/N_{\text{ISO}}}^2 + \sigma_{\text{Bias}}^2 \quad (\text{B-22})$$

where $\sigma_{\text{SYS, Blank}}^2 (R_{232})$ was determined with Eq. B-13, $\sigma_{\text{count}/N_{\text{ISO}}}^2$ is the variance of the average count ratio as calculated from counting statistics for the individual measured count ratios, and σ_{Bias}^2 was assigned the value $(0.031 R_{232})^2$ in agreement with the foregoing discussion. This last term was, by far, the dominant contributor to the total variance. In light of our now including an estimate of the bias associated with R_{232} in our error propagation, the variance of R_{232} , as calculated from Eq. B-22, probably overestimates the actual variance by a considerable margin. However, since the discrepancy is on the conservative side, and since use of this variance does not reflect poorly on the ANL results, we recommend that the uncertainties reported with the EOL results be retained.

C. Estimation of Errors in Uranium Isotope Abundances

Expressions describing the relationship between errors in the uranium isotope abundances and errors in the measured uranium isotope ratios were derived by Taylor-series expansion of Eqs. B-1a and B-1b. Because project error requirements were not stipulated in terms of individual abundance determinations

*S. K. Aggarwal, S. B. Manohar, S. N. Acharya, S. Prakash, and H. C. Jain, Phys. Rev. C, **20**, 1533 (1979).

Fig. B-1. Difference Between $^{232}\text{U}/^{233}\text{U}$ Isotope Ratios as Measured by Mass Spectrometry and Alpha Counting as a Function of the Alpha-Counting Ratio

but in terms of the sum of the fissile isotopes, ^{233}U and ^{235}U , expressions relating errors in this sum to errors in the measured ratios were also employed by the report-generator computer program. In the following subsections, we present the expressions that were used for calculating the quoted uncertainty in the individual weight percent abundance results. Bias error in these results is treated immediately below.

1. Estimation of Bias in Uranium Isotopic Abundances

From Taylor-series expansion of Eq. B-1a, the percent relative bias in WPC_{233} is given by

$$B_R(\text{WPC}_{233}) = -\sum_i [f_i B_R(R_i)] \quad (\text{B-23})$$

where $B_R(Y)$ stands for the percent relative bias in the variable Y , and f_i is the weight fraction abundance of isotope i in the uranium sample and is equal to WPC_i divided by 100%. Similarly, the percent relative bias for the abundance of each of the other isotopes, from Eq. B-1b, is

$$B_R(\text{WPC}_i) = B_R(R_i) - \sum_j [f_j B_R(R_j)] \quad (\text{B-24})$$

The variables $B_R(R_i)$ have been evaluated previously (cf. Eq. B-6 and discussion of bias error in APHA ratio measurement). These expressions indicate that the magnitude of the bias associated with each measured isotope abundance depends upon the isotopic composition of the uranium being analyzed. In addition, there is a correlation among the $B_R(R_j)$ values for ratios measured by TIMS, since bias errors in these ratios share a common source and have the same sign.

A limit to probable bias in the WPC_{233} values reported by ANL can be established by equating the values for f_i in Eq. B-23 with those for a hypothetical uranium sample that is enriched in each minor isotope (i.e., each isotope other than ^{233}U) to a level consistent with each isotope's maximum enrichment among the LWBR fuel rod segments. Such a uranium sample might contain, for example, 15% ^{234}U , 7% ^{235}U , 2% ^{236}U , 1% ^{238}U , and 0.3% ^{232}U . Substituting these values together with the appropriate $B_R(R_j)$ into Eq. B-23 and choosing signs for each $B_R(R_j)$ to give the largest $B_R(\text{WPC}_{233})$ within the constraints of the correlation among some $B_R(R_i)$, one obtains

$$\begin{aligned} B_R(\text{WPC}_{233})|_{\max} &= [(0.15)(0.017) + (0.07)(0.033) + (0.02)(0.05) \\ &\quad + (0.01)(0.083) \pm (0.003)(4.0) = 0.0189\% \end{aligned} \quad (\text{B-25})$$

For each of the other uranium isotopes, a corresponding maximum potential bias may be estimated from Eq. B-24:

$$\begin{aligned}
B_R(WPC_{232})|_{\max} &= \pm 4.0 - 0.0069 \pm 0.012 = 4.0\% \\
B_R(WPC_{234})|_{\max} &= 0.017 - 0.0069 \pm 0.012 = 0.022\% \\
B_R(WPC_{235})|_{\max} &= 0.033 - 0.0069 \pm 0.012 = 0.038\% \\
B_R(WPC_{236})|_{\max} &= 0.05 - 0.0069 \pm 0.012 = 0.055\% \\
B_R(WPC_{238})|_{\max} &= 0.083 - 0.0069 \pm 0.012 = 0.088\%
\end{aligned} \tag{B-26}$$

For the sum of the abundances of the fissile isotopes, $^{233}\text{U} + ^{235}\text{U}$, defined for the following treatment as WPC_{FISS} , the percent relative bias is given by the expression

$$B_R(WPC_{\text{FISS}}) = B_R(WPC_{233}) + \left(\frac{WPC_{235}}{WPC_{\text{FISS}}} \right) B_R(R_{235}) \tag{B-27}$$

Thus, the magnitude of the potential bias in this result also will depend on the specific isotopic composition of the uranium in the sample and will be larger for segments that are less highly enriched in ^{233}U . The maximum probable bias associated with WPC_{FISS} for the LWBR fuel rod segments may be estimated using this expression and the same hypothetical, worst-case composition that we used above. For this composition, WPC_{FISS} is calculated to be 81.7%. Making appropriate substitutions into Eq. B-27, we find

$$B_R(WPC_{\text{FISS}})|_{\max} = 0.0189\% - \left(\frac{7\%}{81.7\%} \right) 0.038\% = 0.016\% \tag{B-28}$$

In this calculation, the fact that $B_R(WPC_{233})$ and $B_R(R_{235})$ have opposite signs is taken into account. This exercise demonstrates that potential bias in the sum of the abundances of fissile isotopes for all LWBR fuel rod segments analyzed at ANL will be less than 0.016%, relative. It follows, therefore, that the limit (0.05%) to allowable bias in these results, as specified in the project error requirements, was not exceeded for any individual segment. Thus, this requirement is shown to have been met.

2. Estimation of Standard Deviation of Reported Uranium Isotopic Abundances

From Taylor-series expansion of Eq. B-1a, the absolute variance, $\sigma^2(WPC_i)$, of each uranium isotope abundance in a particular sample may be estimated from the absolute variance of the measured ratios according to the following expressions:

$$\begin{aligned}
\sigma^2(WPC_{233}) &= WPC_{233}^2 f_{233}^2 \sum_j \sigma^2(R_j), \quad i = 233 \\
\sigma^2(WPC_i) &= WPC_{233}^2 [(1 - 2f_i)\sigma^2(R_i) + f_i^2 \sum_j \sigma^2(R_j)], \quad i \neq 233
\end{aligned} \tag{B-29}$$

where the summations are taken over all measured isotope ratios R_j ($j = 232, 234, 235, 236, \text{ and } 238$). For the sum of the fissile isotopes, the corresponding expression is:

$$\sigma^2(\text{WPC}_{233} + \text{WPC}_{235}) = \text{WPC}_{233}^2 [(1 - 2f_{\text{FISS}}) \sigma^2(R_{235}) + f_{\text{FISS}}^2 \sum \sigma^2(R_j)] \quad (\text{B-30})$$

where f_{FISS} stands for the weight fraction of fissile isotopes in the uranium sample and is given by WPC_{FISS} divided by 100%. Using these expressions and the variances [i.e., $\sigma^2(R_j)$] for the individual measured ratios, as derived earlier, the report-generator program calculated a variance for each individual weight-percent abundance value that was reported during the EOL campaign. On the basis of the treatment just presented, we believe that these estimated variances represent a thorough and realistic measure of the random and randomly varying systematic errors affecting the weight-percent abundance results.

To determine whether the project error requirement on the standard deviation of the sum of fissile isotope abundances was met, we need only examine the uncertainties quoted for these results, given in Table B-3. These data show that, except for two low-uranium segments (D-01 and R-01), which showed % RSD's between 0.08% and 0.09%, the % RSD of this sum did not exceed 0.03%. This value is smaller than the limit (0.08%) to allowable error specified by the project error requirements. For the two low-U segments, the appropriate limit to allowable error is given by $0.08\% / \text{TU}_{\text{seg}}$ (cf. Table 2 in the body of the report). Hence, the % RSDs estimated for these segments also fall within the limit to allowable error. This requirement was thus met for each and every rod segment.

Table B-3. Estimated Standard Deviations of the Fissile Uranium Isotope Abundance Sums for Individual LWBR Fuel Rod Segments

Rod No.	Estimated Relative Standard Deviation for Indicated Segment, %														
	01	02	03	04	05	06	07	08	09	10	11	12	13	14	15
B	0.015	0.004	0.005	0.006	0.005	0.004	0.011	--	--	--	--	--	--	--	--
C	0.024	0.004	0.004	0.005	0.005	0.005	0.013	--	--	--	--	--	--	--	--
D	0.086 ^a	0.003	0.004	0.004	0.004	0.004	0.028	--	--	--	--	--	--	--	--
E	0.017	0.006	0.006	0.007	0.008	0.007	0.007	--	--	--	--	--	--	--	--
F	0.026	0.004	0.005	0.006	0.006	0.007	0.008	0.008	--	--	--	--	--	--	--
G	0.011	0.003	0.005	0.005	0.005	0.008	0.006	--	--	--	--	--	--	--	--
H	0.028	0.008	0.006	0.005	0.005	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.017
I	0.010	0.005	0.006	0.007	0.007	0.007	0.004	0.006	--	--	--	--	--	--	--
J	0.008	0.006	0.008	0.009	0.010	0.009	0.006	0.006	--	--	--	--	--	--	--
K	0.010	0.004	0.005	0.006	0.007	0.006	0.008	0.009	--	--	--	--	--	--	--
L	0.012	0.005	0.006	0.007	0.007	0.008	0.006	0.006	--	--	--	--	--	--	--
M	0.008	0.005	0.007	0.006	0.003	0.013	--	--	--	--	--	--	--	--	--
N	0.009	0.004	0.006	0.005	0.005	0.015	--	--	--	--	--	--	--	--	--
O	0.005	0.009	0.010	0.007	0.006	0.014	--	--	--	--	--	--	--	--	--
P	0.006	0.009	0.008	0.007	0.006	0.015	--	--	--	--	--	--	--	--	--
Q	0.007	0.008	0.008	0.007	0.005	0.016	--	--	--	--	--	--	--	--	--
R	0.088 ^a	0.012	0.006	0.006	0.006	0.005	0.006	0.007	0.006	0.006	0.006	0.006	0.007	0.007	0.025

^aLow-U Sements: D-01 contained 0.10930 g U total, allowable % RSD = 0.091%.
R-01 contained 0.05101 g U total, allowable % RSD = 0.196%.

APPENDIX C

ISOMASS 54R CALIBRATION DATA

Calibration of the Isomass 54R Thermal Ionization Mass Spectrometer, which was used in measuring uranium isotope ratios during the EOL campaign, was described in Appendix B. In this appendix, we report data obtained with the NBS U-500 uranium isotopic standard to validate calibration of the instrument. In acquiring these data, the U-500 standard was analyzed as an unknown, yielding ratios corrected for fractionation bias in the same way that ratios from the segment isotopic and assay samples were corrected. The fractionation factor employed in making the corrections was determined from measured $^{235}\text{U}/^{238}\text{U}$ ratios obtained during the preceding 10 standard analyses; standards other than U-500 were included among these calibration/verification runs, but are not reported.

STANDARD Date	U-500 234/238	Atom Ratios 236/238	235/238	Re
10-22-85	.010417	.001524	.999477	1
10-14-85	.010427	.001524	1.000120	1
10-09-85	.010405	.001521	.999279	1
09-16-85	.010423	.001521	.999585	1
09-11-85	.010423	.001521	.999687	1
09-04-85	.010405	.001526	.999381	1
08-29-85	.010424	.001516	1.000240	1
08-20-85	.010429	.001525	1.000190	1
08-20-85	.010423	.001523	.999860	1
08-19-85	.010420	.001523	.999399	
08-13-85	.010428	.001528	.999827	
08-13-85	.010429	.001529	1.000510	
08-12-85	.010419	.001522	.999798	
08-08-85	.010441	.001531	1.000360	
08-08-85	.010416	.001525	.999495	
08-07-85	.010422	.001523	1.000090	
08-07-85	.010402	.001522	.998926	
08-05-85	.010426	.001524	1.000150	14
07-30-85	.010417	.001524	.999588	14
07-19-85	.010413	.001521	.999559	14
07-15-85	.010416	.001522	.999492	14
06-28-85	.010419	.001521	.999183	14
06-12-85	.010415	.001525	.999391	14
06-11-85	.010421	.001523	.999721	14
05-22-85	.010417	.001518	.999945	13
05-20-85	.010411	.001519	.999341	13
04-11-85	.010423	.001523	.999873	13
04-11-85	.010414	.001517	1.000000	13
04-10-85	.010442	.001528	1.000130	13
04-09-85	.010429	.001529	.999568	13
03-20-85	.010437	.001529	1.000010	13
03-18-85	.010431	.001527	1.000450	13
02-28-86	.010426	.001525	1.000090	12
02-26-85	.010411	.001527	.999311	12
02-04-85	.010426	.001529	1.000090	12
02-04-85	.010419	.001518	.999505	12
01-31-85	.010422	.001525	.999815	12
01-30-85	.010432	.001518	1.000300	12
01-15-85	.010416	.001526	.999635	12
01-07-85	.010422	.001525	1.000100	12
12-14-84	.010425	.001523	.999941	12
12-11-84	.010424	.001520	1.000290	11
12-04-84	.010426	.001526	1.000050	11
11-28-84	.010412	.001524	.998930	11
11-26-84	.010413	.001519	.999449	11
11-19-84	.010434	.001523	1.000030	11
11-02-84	.010410	.001522	.999000	11
10-29-84	.010422	.001521	.999667	11
10-04-84	.010416	.001520	.999250	11
09-24-84	.010423	.001520	.999825	11
09-10-84	.010417	.001527	.999744	10
09-07-84	.010412	.001526	.998854	10
09-06-84	.010413	.001522	.999334	10
09-05-84	.010419	.001522	.999235	10

08-31-84	.010421	.001524	.999525	105
08-30-84	.010413	.001520	.999545	104
08-29-84	.010432	.001526	1.000400	103
08-22-84	.010436	.001526	1.000470	102
08-21-84	.010427	.001520	1.000130	99
08-21-84	.010424	.001523	.999997	98
08-17-84	.010423	.001518	.999712	94
08-16-84	.010412	.001520	.999144	91
08-15-84	.010426	.001519	1.000360	88
08-14-84	.010429	.001521	1.000750	85
08-13-84	.010434	.001521	1.000560	83
05-14-84	.010415	.001515	.999728	81
05-14-84	.010421	.001524	.999879	80
05-11-84	.010426	.001519	1.000150	79
05-11-84	.010409	.001523	.999362	78
05-10-84	.010411	.001516	.999045	77
05-10-84	.010405	.001519	.998502	76
01-03-84	.010417	.001523	.999504	73
11-07-83	.010411	.001526	.999684	66
10-24-83	.010421	.001524	.999847	63
10-24-83	.010427	.001524	1.000370	62
10-10-83	.010429	.001526	1.000450	61
09-01-83	.010420	.001524	.999922	60
06-24-83	.010427	.001523	.999757	57
05-03-83	.010428	.001519	.999708	55
03-29-83	.010428	.001522	.999967	37
03-22-83	.010426	.001518	.999769	26
03-21-83	.010428	.001527	.999926	19
03-21-83	.010412	.001516	.999632	18
03-15-83	.010419	.001520	.999678	17
03-15-83	.010420	.001523	.999994	16
02-17-83	.010419	.001519	.999532	13
02-15-83	.010418	.001520	.999210	12
02-07-83	.010422	.001522	.999676	10
12-13-82	.010423	.001527	.999892	9
12-03-82	.010412	.001513	.999013	8
12-03-82	.010418	.001520	.999568	7
12-03-82	.010422	.001520	.999766	6
12-03-82	.010422	.001518	.999269	5
12-02-82	.010428	.001523	1.000310	4
12-02-82	.010417	.001519	.999517	3
12-02-82	.010421	.001516	.999947	2

AVERAGE	.010421	.001522	.999752
Std dev	.000008	.000004	.000432
RSD, %	.074923	.231861	.043213

NBS VALUES	.010422	.001519	.999700

DEV FROM NBS	-.000001	.000003	.000052

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APPENDIX D

ESTIMATION OF BIAS AND STANDARD DEVIATION FOR SEGMENT URANIUM ASSAY RESULTS

A. Introduction

The total weight of uranium in each fuel rod segment was computed in two different ways with data obtained during the segment's dissolution and during subsequent analysis of the solution samples taken from the dissolver system. The result of one of these calculations, designated the 950a-MSID assay result, was based largely on the MSID assay of uranium in the blend tank solution spiked with NBS 950a, a natural uranium standard containing primarily ^{238}U . The other result, designated the 993-MSID assay result, was largely based on the uranium concentration in the unspiked blend tank (UBT), which was determined by spiking aliquots of the UBT solution with NBS 993, a ^{235}U standard. The 950a-MSID assay result, together with the sum of the fissile isotope abundances determined for the corresponding segment, was also used to calculate the segment fissile uranium content.

Each of these assay results was calculated using a computer program that compiled data from various operations within the project and combined these data in the manner required by the calculations. This program also statistically analyzed replicate measurement data, where available, and estimated the uncertainty associated with each segment uranium assay result. This quoted uncertainty represented an estimate of the standard deviation of each individual result and included contributions from both random errors and randomly varying systematic errors in the measurements that were made in carrying out the uranium assays. Potential bias errors in the reported assay results were not explicitly quoted in the computer-generated reports on ANL's EOL work, since they were characterized in a pre-EOL error analysis and shown to be small enough to meet EOL requirements under normal operating conditions.

In this appendix, we develop estimates of the potential bias associated with the individual uranium assay results and also derive the expressions that were used by the report-generator computer program in estimating the quoted standard deviations. When appropriate, we present experimental data to support the validity of assumptions involved in our treatment of these errors.

B. Bias in the 950a-MSID Assay Result

The total segment uranium represented by the 950a-MSID assay result, ${}^T\text{U}_{\text{seg}, 950}$, was calculated as the quantity of uranium in the spiked blend tank (U_{SBT}) plus the (small) quantities that were removed from the dissolver system as second-dissolution (DS) and unspiked blend tank (UBT) samples. That is,

$${}^T\text{U}_{\text{seg}, 950} = \text{U}_{\text{SDS}} + \text{U}_{\text{UBTS}} + \text{U}_{\text{SBT}} \quad (\text{D-1})$$

where U_{SDS} is the total weight of uranium in the DS samples, and U_{UBTS} is the corresponding total weight of U in the UBT samples. Bias in ${}^T\text{U}_{\text{seg}, 950}$ is given by the sum of the biases in each of these three terms:

$$B(^T U_{\text{seg},950}) = B(U_{\text{SDS}}) + B(U_{\text{UBTS}}) + B(U_{\text{SBT}}) \quad (\text{D-2})$$

Here, the notation $B(X)$ stands for the bias in the variable X . This relationship may also be expressed in terms of percent relative bias, designated $B_R(X)$:

$$\begin{aligned} B_R(^T U_{\text{seg},950}) &= \left(\frac{U_{\text{SDS}}}{^T U_{\text{seg},950}} \right) B_R(U_{\text{SDS}}) + \left(\frac{U_{\text{UBTS}}}{^T U_{\text{seg},950}} \right) B_R(U_{\text{UBTS}}) \\ &+ \left(\frac{U_{\text{SBT}}}{^T U_{\text{seg},950}} \right) B_R(U_{\text{SBT}}) \end{aligned} \quad (\text{D-3})$$

To evaluate the bias in $^T U_{\text{seg},950}$ from either Eq. D-2 or D-3, one must first evaluate the contributing bias terms. We estimate the magnitude of each term separately in Sections 1, 2, and 3, below, and combine them in Section 4 to estimate $B_R(^T U_{\text{seg},950})$.

1. Bias in U_{SDS}

In computing $^T U_{\text{seg},950}$, we calculated U_{SDS} as the product of the total weight of solution contained in the DS samples, W_{SDS} , and the weight concentration of uranium in the DS solution, $[U]_{\text{DS}}$:

$$U_{\text{SDS}} = W_{\text{SDS}} [U]_{\text{DS}} \quad (\text{D-4})$$

The percent relative bias in U_{SDS} may thus be written in terms of these variables as

$$B_R(U_{\text{SDS}}) = B_R(W_{\text{SDS}}) + B_R([U]_{\text{DS}}) \quad (\text{D-5})$$

The weight of solution in the DS samples was determined by weighing the bottles that contained the solution samples and summing the net solution weights. Practically speaking, relative bias in the solution weight is negligibly small because the balance used for these weighings was frequently calibrated with NBS-traceable standard weights.

The uranium concentration, $[U]_{\text{DS}}$, was calculated from weight data and thermal ionization mass spectrometry (TIMS) data associated with the MSID assay of the DS solution. Bias in $[U]_{\text{DS}}$ is, therefore, best estimated by propagation of bias errors in the individual measurements that enter into this calculation. The expression used in calculating $[U]_{\text{DS}}$ is

$$[U]_{\text{DS}} = \left(\frac{{}^{993}\text{W}_{\text{SP}}}{\text{DSW}_{\text{ALQ}}} \right) \left(\frac{{}^{993}\text{f}_{235} \text{DSR}_{3/5}}{\text{segf}_{233} - \text{segf}_{235} \text{DSR}_{3/5}} \right) \quad (\text{D-6})$$

where $^{993}W_{SP}$ is the weight of NBS 993 in the spike that was mixed with the aliquot, of weight W_{ALQ} , from the DS solution sample; the f variables are weight fractions of the subscripted isotope in the superscripted material; and $^{DS}R_{3/5}$ is the corrected weight ratio of ^{233}U to ^{235}U in the spiked aliquot, as measured by TIMS. Noting that the second term in the denominator of Eq. D-6 (i.e., $f_{235}^{DS}R_{3/5}$) is much smaller than the first term, at least for the purpose of our error propagation, and expanding Eq. D-6 in a Taylor series, one may obtain the following expression for the relative bias in $[U]_{DS}$:

$$B_R([U]_{DS}) = B_R(^{993}W_{SP}) + B_R(^{993}f_{235}) + B_R(^{DS}R_{3/5}) - B_R(^{DS}W_{ALQ}) - B_R(^{seg}f_{233}) \quad (D-7)$$

Each of the terms on the right side of this expression may be evaluated as follows.

a. Bias in the Weight of NBS 993 Spike, $B_R(^{993}W_{SP})$

The NBS 993 spikes used in the EOL campaign were prepared batchwise by dispensing weighed aliquots of a stock solution prepared for each batch (approximately 100 spikes) from an individual vial of NBS 993. Bias errors from the weighing of aliquots were rendered negligible by calibration of the balance used for the weighings with NBS-traceable standard weights. Hence, bias errors in the spike weights arose primarily from errors in defining the stock solution concentration.

For the EOL campaign, BAPL specified that ANL apply bias corrections to assay results based on NBS 993 so as to accommodate differences between the NBS 993 and NBS 950a standards, since NBS 950a was designated as the primary uranium assay standard for the project. In keeping with this specification, we cross-standardized each batch of NBS 993 spikes to NBS 950a by adding four weighed aliquots from each stock solution to pre-weighed NBS 950a spikes and determining the stock solution concentration by MSID. Results of these cross-standardizations are shown in Table D-1. Over the nine batches of NBS 993 spikes that were prepared for the POB effort, the measured weight of uranium in the spikes exceeded the weight calculated from information on the NBS 993 certificate by an average of 0.044%, relative. We could not determine from the data at hand whether this difference reflected an actual difference between the standards, or whether it arose from errors introduced during preparation of the NBS 993 spikes. Regardless of the cause, the data show that separate standardization of each spike batch was appropriate to minimize differences between assays based on the two standards.

Residual error in the NBS 993 spike weights, after cross-standardization, may be identified with (1) uncertainty in the certified assay value for NBS 950a and (2) uncertainty in the corrections applied on the basis of the four measurements that were performed on each batch. For the purpose of our analysis of errors in the EOL uranium assays, this latter uncertainty is treated as a randomly varying systematic error since, although it has a given magnitude and direction for each individual batch of spikes, its magnitude and direction may be presumed to vary randomly among the different batches.

Table D-1. Results from Cross-Standardization of NBS 993 Spike Batches with NBS 950a

NBS 993 Batch Desig.	NBS 993 Vial No.	No. of Spikes in Batch	Average Difference Between Measured & NBS Values, % Rel.	% RSD from Four MSID Measurements
93A	70	98	+0.062	0.024
93B	128	98	+0.068	0.020
93C	93	99	+0.026	0.008
93D	126	98	+0.052	0.015
93E	127	99	+0.042	0.020
93F	124	98	+0.068	0.018
93G	111	99	+0.018	0.020
93H	107	100	+0.025	0.026
93I	106	98	+0.054	0.014
Average % Rel. Difference: +0.044% (% RSD = 0.020%)				
Pooled % RSD of Individual Measurement: 0.019%				

In contrast, error in the NBS 950a assay value affects all batches equally and, therefore, represents a residual bias error in the corrected spike weights. From the NBS certificate for NBS 950a, this uncertainty does not exceed 0.02%, relative, at a confidence level of at least 95%. Taking 0.01%, relative, as a reasonable estimate of the one-sigma uncertainty in the NBS 950a assay value, we assign the value 0.01% to $B_R(^{993}\text{W}_{\text{SP}})$.

Because this assignment requires that we assume elimination, by our calibration procedure, of any bias that existed between the NBS 993 and NBS 950a standards, we tested the procedure's effectiveness in a short pre-EOL study. For this test, six weighed portions of NBS 950a uranium were mixed with previously cross-standardized NBS 993 spikes, and the NBS 950a uranium was determined as an unknown by MSID. The results of these determinations are shown in Table D-2. The average percent relative difference between the weighed and measured amounts of NBS 950a was -0.009%, with a % RSD for the difference of 0.012%. These results provide no evidence of a significant residual

Table D-2. Confirmation of NBS 993 Spike Calibration vs. NBS 950a

993 Spike No.	NBS 950a Added, mg U	NBS 950a Found, mg U	Rel. Difference, %
35380	0.49862	0.49849	-0.026
35381	0.50399	0.50394	-0.010
35382	1.02071	1.02081	+0.010
35383	1.01704	1.01696	-0.008
35384	2.05426	2.05416	-0.005
35385	2.02065	2.02034	-0.015
Average:			-0.009
Std. Dev. of Rel. Diff:			0.012

bias between the two standards. Thus, the value we assign to $B_R(^{993}W_{SP})$ is a legitimate estimate of potential bias in the NBS 993 spike weights.

b. Bias in the Weight Fraction of ^{235}U in NBS 993, $B_R(^{993}f_{235})$

The NBS certificate for NBS 993 specifies the ^{235}U content of this material as 99.8195 ± 0.0013 at %. Since the uncertainty in this value translates almost directly into the uncertainty in $^{993}f_{235}$, we assign, on a one-sigma basis, a value of 0.0007%, relative, to $B_R(^{993}f_{235})$.

c. Bias in the Measured Ratio of ^{233}U to ^{235}U in the Spiked Aliquot, $B_R(^{DS}R_{3/5})$

Potential bias in uranium isotope ratios measured by TIMS was discussed in detail in Appendix B. The only significant bias errors in these measured ratios stem from uncertainty in the uranium isotopic standard used for calibrating the TIMS instrument. From Eq. B-5 in that Appendix, $B_R(^{DS}R_{3/5})$ is estimated to be 0.033%.

d. Bias in the Weight of DS Solution Spiked with NBS 993, $B_R(^{DS}W_{ALQ})$

Typically, a 10-g aliquot of the DS solution sample was weighed to the nearest 0.1 mg on a balance calibrated with NBS-traceable standard weights. Bias errors associated with this weighing are thus negligible relative to other terms in Eq. D-7.

e. Bias in the Weight Fraction of ^{233}U in the Segment Uranium, $B_R(^{seg}f_{233})$

The weight percent abundance of ^{233}U in uranium from each EOL segment was determined as part of the ANL segment analysis. Relative errors in the weight fraction of a given isotope are identical to those in the weight percent abundance of that isotope, as was shown in Appendix B. Hence, $B_R(^{seg}f_{233})$ can be shown to be dependent on the exact isotopic composition of the segment uranium and to fall in the range 0 to 0.019% for all segments processed during the EOL campaign (see Eq. B-25 of Appendix B). It should be noted that $B_R(^{DS}R_{3/5})$ and $B_R(^{seg}f_{233})$ derive predominantly from a common source, uncertainty in the U-500 isotopic standard; thus, both these bias terms always have the same sign regardless of their respective magnitudes.

f. Calculation of $B_R(U_{DS})$

Substituting the foregoing estimates of the magnitudes of the various contributors to bias in $[U]_{DS}$ into Eq. D-7 gives

$$B_R([U]_{DS})|_{\max} = (0.01\%) + (0.0007\%) \\ + (0.033\%) - (0.00\%) - (0.00\%) = 0.044\% \quad (D-8)$$

We chose $B_R(^{seg}f_{233}) = 0$ and the signs of the other bias terms to give the maximum value for the sum. Since $B_R(W_{SDS})$ was judged earlier to be negligibly small, we may now write, for the maximum probable bias in U_{SDS} , using Eq. D-5,

$$B_R(U_{SDS})|_{\max} = B_R([U]_{DS}) = 0.044\% \quad (D-9)$$

2. Bias in U_{UBTS}

In calculating $TU_{seg,950}$, we computed U_{UBTS} in a manner analogous to that used for U_{SDS} . Thus, U_{UBTS} was calculated as the product of the weight of solution removed from the UBT as samples and the weight concentration of segment uranium in the UBT solution:

$$U_{UBTS} = W_{UBTS} [U]_{UBT} \quad (D-10)$$

The ways in which the solution weight and concentration values for the UBT samples were obtained likewise mirrored those for the DS samples, with two exceptions: (1) the $^{233}\text{U}/^{235}\text{U}$ ratio in the spiked aliquot of the UBT solution was determined using a unique, internal-standard procedure developed at ANL for the TIMS measurements, and (2) the uranium concentration was measured on single aliquots from each of two separate UBT solution samples rather than for one sample only, as in the DS case. Neither of these differences affects the magnitude of the bias errors associated with the measurements. The internal-standard procedure for TIMS ratio measurements improves the precision of the measurement but is still calibrated vs. NBS U-500 isotopic standard. Replication of measurements also improves precision but has no effect on systematic errors, such as a bias in the results.

Based on these considerations, it is clear that the percent relative bias in U_{UBTS} should be identical to that in U_{SDS} for any given segment. Thus, we assign the same value for the maximum potential bias in U_{UBTS} as we derived above for U_{SDS} :

$$B_R(U_{UBTS})|_{\max} = 0.044\% \quad (D-11)$$

3. Bias in U_{SBT}

The value of U_{SBT} for each segment depends on the weight of the NBS 950a spike added to the blend tank solution ($^{950}W_{SP}$) and the ratio of ^{233}U to ^{238}U in the SBT solution ($^{SBT}R_{3/8}$), as measured by TIMS. This value was calculated using the relationship

$$U_{SBT} = ^{950}W_{SP} \left(\frac{^{950}f_{238} \text{ } ^{SBT}R_{3/8}}{^{seg}f_{233} - ^{seg}f_{238} \text{ } ^{SBT}R_{3/8}} \right) \quad (D-12)$$

Here, the individual variables are defined in a manner analogous to those in Eq. D-6. We applied a treatment like that used in estimating $B_R([U]_{DS})$ to obtain the relationship between $B_R(U_{SBT})$ and the biases in the other variables. Thus,

$$B_R(U_{SBT}) = B_R(^{950}W_{SP}) + B_R(^{950}f_{238}) \\ + B_R(^{SBT}R_{3/8}) - B_R(^{seg}f_{233}) \quad (D-13)$$

The individual terms in this expression may be evaluated in the same manner as were the corresponding terms in Eq. D-6. From the discussion of bias in the NBS 993 spikes in Section B.1.a above, we assign the value 0.01% to $B_R(^{950}W_{SP})$. Since the enrichment of ^{238}U in NBS 950a (99.2844% by weight) is comparable to the enrichment of ^{235}U in NBS 993, we take $B_R(^{950}f_{238})$ to be about the same as $B_R(^{993}f_{235})$, or 0.001%. From Eq. B-5 in Appendix B, $B_R(^{SBT}R_{3/8})$ is estimated to be 0.083%. Bias in $^{seg}f_{233}$ is the same for this expression as for Eq. D-6; thus, from our earlier discussion of this error, we assign the value zero to $B_R(^{seg}f_{233})$ to maximize our estimate of potential bias in U_{SBT} . With these estimates in hand, we may now write

$$B_R(U_{SBT})|_{\max} = 0.01\% + 0.001\% + 0.083\% - 0\% = 0.094\% \quad (D-14)$$

4. Evaluation of $B_R(^T U_{seg,950})$

To calculate a value for $B_R(^T U_{seg,950})$ using Eq. D-3, one must have not only estimates of the $B_R(X)$ in the expression but also the fractions of $^T U_{seg,950}$ that U_{SDS} , U_{UBTS} , and U_{SBT} represent. Typically, the DS samples contained only about 0.1% of the total uranium for the dissolved segment, the UBT samples contained 2 to 3%, and the SBT accounted for the remainder. Because the value of $B_R(^T U_{seg,950})$ is not very sensitive to the distribution of uranium among these locations, as long as the actual distribution does not vary too drastically from the norm, we assume, for the present calculation, a distribution corresponding to these typical values. Thus, substituting for the variables in Eq. D-3, we get

$$B_R(^T U_{seg,950})|_{\max} = 0.001(0.044\%) + 0.025(0.044\%) \\ + 0.974(0.094\%) = 0.093\% \quad (D-15)$$

This estimate of the maximum potential bias in $^T U_{seg,950}$ includes only contributions from potential errors in the measurements performed in evaluating this uranium assay result. It does not include potential bias errors due to fuel losses that might have occurred during dissolution of the segments in preparation for the measurements. Such errors are discussed in Section V.C of the report body. Under conditions of normal dissolver operation, fuel losses during dissolution amounted to no more than about 0.03%, relative. For segments dissolved under such conditions, then, a limit to potential bias in the 950a-MSID assay result may be set at 0.12%, relative, by adding the potential bias associated with our measurements and that from potentially lost fuel; the uranium assays for these

segments thus met the project requirement that this bias be less than 0.15%, relative.

For eight of the 135 fuel-bearing segments that were processed in the EOL campaign, there were mishaps during dissolver operations which increased the potential for losing segment fuel (see Section V.C.3.b of the report body). When the measurement bias and fuel-loss bias are combined for these segments as shown in Table D-3, potential bias in the 950a-MSID results for five of them exceeds the limit (0.15%) to allowable bias as established in the project error requirements. However, the project error limits were defined such that they would propagate to the limits (also 0.15%) established in the DOE-NR parametric requirements on bias in determining the uranium loading for the whole rod (cf. Table 1 in the report body). Bias in the segment uranium assays propagates to that in the whole-rod loading through the sum of the segment assay results. Thus, if the potential bias in the sum of segment assay results is smaller than the value that would accrue if all segments in a rod showed a potential bias equal to the allowed limit, the segment assays would satisfy the intent, if not the letter, of the project requirements. Bias in the sum of segment assay results for each rod was calculated by summing the maximum probable bias estimates for the $^{235}\text{U}_{\text{seg},950}$ results from the individual segments in the rod; results of these calculations are shown in Table D-4. Because each segment having a potential bias greater than the limit prescribed by project error requirements contained only a small fraction of the rod's total uranium, the mishaps with these segments only increased the potential bias in the sum for each corresponding rod by 0.03%, relative, or less. Thus, the effect of each mishap on the sum of segment assay results was quite small, and the magnitude of potential bias errors in ANL's uranium assays is acceptable.

Table D-3. Potential Bias in 950a-MSID Uranium Assay Results

Segment No.	Maximum Probable Bias in Result, % Relative		
	From Fuel Losses ^a	From Measurements	Total
H-08	0.13	0.09	0.22 ^b
J-03	0.15	0.09	0.24 ^b
J-08	0.07	0.09	0.18 ^b
N-02	0.03	0.09	0.12
N-06	0.09	0.09	0.18 ^b
O-03	0.25	0.09	0.34 ^b
All Others	0.03	0.09	0.12

^aData from Table 20 in report body.

^bResult fails to meet project requirements on bias in segment uranium assay.

Table D-4. Potential Bias in Sums of Segment Uranium Assay Results Based on 950a-MSID

Rod No.	Maximum Probable Bias in Sum of Segment Uranium Assay Results, % Relative
H	0.13
J	0.14
N	0.12
O	0.15
All Others	0.12

C. Bias in the 993-MSID Assay Result

The 993-MSID uranium assay result was determined as a corroborative check on the 950a-MSID value. It was calculated from the relationship

$${}^T U_{\text{seg},993} = U_{\text{SDS}} + U_{\text{UBT}} = U_{\text{SDS}} + W_{\text{UBT}} [U]_{\text{UBT}} \quad (\text{D-16})$$

where W_{UBT} stands for the total weight of solution present in the UBT before sampling, and the other variables are as defined earlier. Bias in this assay result may be estimated from potential biases in the individual terms, as was done for the 950a-MSID result. In this case, the appropriate expression is

$$\begin{aligned} B_R({}^T U_{\text{seg},993}) &= \left(\frac{U_{\text{SDS}}}{{}^T U_{\text{seg},993}} \right) B_R(U_{\text{SDS}}) \\ &+ \left(\frac{U_{\text{UBT}}}{{}^T U_{\text{seg},993}} \right) [B_R(W_{\text{UBT}}) + B_R([U]_{\text{UBT}})] \end{aligned} \quad (\text{D-17})$$

Taking $B_R(W_{\text{UBT}})$ to correspond to the resolution (± 1 g) of the balance used in weighing the blend tank assembly relative to W_{UBT} (~ 5 kg or more), we set $B_R(W_{\text{UBT}})$ equal to 0.02%. The other bias terms in Eq. D-17 were established in the previous section. Typically, U_{SDS} accounted for only about 0.1% of the total segment uranium, as we pointed out earlier, so that U_{UBT} may be taken to correspond to 99.9% of the total U. Making appropriate substitutions in Eq. D-17, one finds that

$$B_R({}^T U_{\text{seg},993}) = 0.001(0.044\%) + 0.999(0.02\% + 0.044\%) = 0.064\% \quad (\text{D-18})$$

This result indicates that the potential for bias error in the 993-MSID assay result (maximum probable bias, 0.064%) is somewhat smaller than that for bias error in the 950a-MSID assay result (maximum probable bias, 0.093%). However, it should not be interpreted as an indication of greater overall reliability of the 993-MSID results because, as shown in Section F, below, the precision of the 993-MSID assay is not very high. Because of its high precision and its direct use of NBS 950a as a primary standard, the 950a-MSID assay was designated the primary assay, while the 993-MSID results were used only to corroborate it. Results from these two assay methods, as obtained during the EOL campaign, are discussed in Section G, below, after expressions describing random error in each result are derived.

D. Bias in Determining Segment Fissile Uranium Loading

Potential bias in the determination of fissile uranium loading for each segment is of special interest because the DOE-NR parametric requirements assign a limit of 0.15%, relative, to the bias error permitted for determination of the fissile uranium loading for the whole rod (see Footnote b to Table 1 of the report body). The whole-rod value is computed as the sum of the results for segments from the rod, corrected for shearing losses. To determine whether this requirement was met by ANL's EOL data, a realistic estimate of the maximum probable bias associated with these results had to be made. We derive this estimate in the following paragraphs, using approximate relationships obtained from and supported by our earlier discussions.

The fissile uranium loading of each LWBR rod segment was computed as the product of the 950a-MSID uranium assay result for the segment and the weight fraction of fissile isotopes ($^{233}\text{U} + ^{235}\text{U}$) in the segment uranium. Relative bias in this product is given by the sum

$$B_R(^T\text{U}_{\text{seg,FISS}}) = B_R(^T\text{U}_{\text{seg,950}}) + B_R(^{\text{seg}}f_{\text{FISS}}) \quad (\text{D-19})$$

where the individual terms are defined in a manner consistent with earlier notation.

To simplify the expressions involved in deriving $B_R(^T\text{U}_{\text{seg,FISS}})$, we make use of the near identity between $B_R(^T\text{U}_{\text{seg,950}})$ and $B_R(\text{U}_{\text{SBT}})$, as indicated in Eqs. D-14 and D-15. Using this relationship and the approximate expression in Eq. D-13 to describe $B_R(\text{U}_{\text{SBT}})$, we may write

$$B_R(^T\text{U}_{\text{seg,950}}) \simeq B_R(^{950}\text{W}_{\text{SP}}) + B_R(^{950}f_{238}) + B_R(^{\text{SBT}}\text{R}_{3/8}) - B_R(^{\text{seg}}f_{233}) \quad (\text{D-20})$$

Substituting Eq. D-20 into Eq. D-19 together with the expression for $B_R(\text{WPC}_{\text{FISS}})$ from Eq. B-27 of Appendix B, and noting that $B_R(^{\text{seg}}f_{233}) = B_R(\text{WPC}_{233})$ and $B_R(^{\text{seg}}f_{\text{FISS}}) = B_R(\text{WPC}_{\text{FISS}})$ we obtain

$$B_R(^{TU}_{seg,FISS}) \approx$$

$$B_R(^{950}W_{SP}) + B_R(^{950}f_{238}) + B_R(^{SBTR}_{3/8}) + \left(\frac{WPC_{235}}{WPC_{FISS}} \right) B_R(R_{235}) \quad (D-21)$$

Because the bias in each ratio measured by TIMS (i.e., $^{SBTR}_{3/8}$ and R_{235}) is related to uncertainty in the certified ratio of the isotopic SRM U-500 through Eq. B-5 in Appendix B, and because $B_R(^{SBTR}_{3/8})$ and $B_R(R_{235})$ always have opposite signs owing to this relationship, we may substitute values for the various terms as in previous discussions and obtain the following estimate of the potential bias in $^{TU}_{seg,FISS}$:

$$B_R(^{TU}_{seg,FISS}) = 0.01\% + 0.001\% + 0.083\% + \left(\frac{WPC_{235}}{WPC_{FISS}} \right) (-0.033\%) \quad (D-22)$$

In the worst-case situation where the ^{235}U content of a particular segment is very small, the last term in this expression approaches zero. Thus, the maximum probable bias in $^{TU}_{seg,FISS}$ is given by the expression

$$B_R(^{TU}_{seg,FISS})|_{\max} = 0.01\% + 0.001\% + 0.083\% - 0\% = 0.094\% \quad (D-23)$$

The identity between Eqs. D-14 and D-23 shows that, when the biases in $^{TU}_{seg,FISS}$ and $^{TU}_{seg,950}$ approach their maximum values, the limiting value is the same. Thus, all conclusions that we reached earlier regarding bias in the 950a-MSID assay results apply with equal validity to bias in $^{TU}_{seg,FISS}$. In particular, the maximum probable bias in the sum of fissile uranium contents determined for segments from any given LWBR fuel rod may be shown not to exceed 0.15%, relative, even after bias contributions from fuel losses during specific segment dissolutions are accounted for (cf. Table D-4). The DOE-NR parametric requirement on bias in the fissile uranium loading for a whole rod should thus be met by ANL's EOL results for this determination.

E. Standard Deviation of the 950a-MSID Uranium Assay Result

Formulas employed in the computer program that estimated uncertainties associated with each uranium assay result were derived by propagation of errors associated with variables that entered into the calculation of the assay results themselves. Using these formulas, the program estimated the standard deviation of each result, tailoring each estimate to the specific conditions that applied for each segment. Derivation of the formula used in estimating the standard deviations of the 950a-MSID assay results is presented here. During the derivation, the relative importance of individual error sources in determining these standard deviations is illustrated by indicating typical values that were encountered for the error contributions from identified sources. The standard deviations estimated for the 950a-MSID assay result from each EOL fuel-bearing segment are given after the way in which these estimates were made is described.

When the weight of uranium in a dissolved segment is computed using Eq. D-1, the variance of $TU_{seg,950}$, designated $\sigma^2(TU_{seg,950})$ consistent with our bias notation, may be expressed as

$$\sigma^2(TU_{seg,950}) = \sigma^2(U_{SDS}) + \sigma^2(U_{UBTS}) + \sigma^2(U_{SBT}) \quad (D-24)$$

This relationship may also be expressed in terms of percent relative standard deviations, $\sigma_R(X)$:

$$\begin{aligned} \sigma_R^2(TU_{seg,950}) &= \left(\frac{U_{SDS}}{TU_{seg,950}} \right)^2 \sigma_R^2(U_{SDS}) + \left(\frac{U_{UBTS}}{TU_{seg,950}} \right)^2 \sigma_R^2(U_{UBTS}) \\ &+ \left(\frac{U_{SBT}}{TU_{seg,950}} \right)^2 \sigma_R^2(U_{SBT}) \end{aligned} \quad (D-25)$$

The individual variance terms in these expressions may be evaluated from the formulas used to compute the values for U_{SDS} , U_{UBTS} , and U_{SBT} in a manner analogous to that used in Section B, above, for estimating the bias in $TU_{seg,950}$. Each variance term is treated individually below. The combination of terms is treated thereafter.

1. Variance in U_{SDS}

From a Taylor-series expansion of Eq. D-4, the variance in U_{SDS} may be related to the variance in W_{SDS} and $[U]_{DS}$:

$$\sigma_R^2(U_{SDS}) = \sigma_R^2(W_{SDS}) + \sigma_R^2([U]_{DS}) \quad (D-26)$$

Errors in weighing solution samples that were taken for each dissolved segment during DDS operations are discussed in Section V.C.3.a.(2) in the report body. The variance of this solution weight was taken as being equivalent to the sum of (1) the random variances associated with balance calibrations and bottle weighings and (2) a systematic variance related to potential losses of the DS solution during the taking of samples. All of these contributing variances were estimated from data obtained during the processing of segments from each individual fuel rod. The random variances were obtained by pooling the variances of replicate weighings, while the systematic variance was calculated as the mean square, over all segments from the rod, of the differences between the net weight of solution in the UBT sample bottles and the apparent change in UBT solution weight on sampling. The computer program determined each of these variances by carrying out the appropriate data manipulations, and then computed $\sigma_R^2(W_{SDS})$ from the relationship

$$\sigma_R^2(W_{SDS}) = \left(\frac{100\%}{W_{SDS}} \right)^2 [\sigma_{\text{Weighing}}^2 + \sigma_{\text{Cal.}}^2 + \sigma_{\text{Loss}}^2] \quad (D-27)$$

where individual variances inside the brackets correspond to the estimated random and systematic variances described above. Values obtained for the bracketed sum of variances from data related to each individual LWBR fuel rod are given in Table V-9 of the report body; the largest of these values corresponds to a % RSD for W_{SDS} of about 2.1%.

Based on similar approximations to those that we employed in estimating $B_R([U]_{SDS})$ (see Eq. D-7), the relative variance in $[U]_{SDS}$ may be expressed as

$$\sigma_R^2([U]_{DS}) = \sigma_R^2(^{993}W_{SP}) + \sigma_R^2(^{DS}W_{ALQ}) + \sigma_R^2(^{DS}R_{3/5}) + \sigma_R^2(^{segf}_{233}) \quad (D-28)$$

Several of the variances in this expression contain both random and systematic components. The systematic variance contributions to each term in Eq. D-28 were individually evaluated from expressions derived as described below. The sum total of the random variance contributions, $\sigma_{R,Rdm}^2([U]_{DS})$, was estimated from the pooled variance of replicate data obtained during processing of segments from each individual fuel rod. Because the uranium concentration of only one DS sample was determined for each segment, such replicate data that related directly to $[U]_{DS}$ were not available. For this reason, it was assumed that the random variance for determination of $[U]_{DS}$ would not differ greatly from the random variance for determining $[U]_{UBT}$, since both determinations involved nearly identical methodology. Thus, the random variance in $[U]_{DS}$ was estimated from the pooled variance, over segments from a given rod, of duplicate $[U]_{UBT}$ determinations that were performed for each segment. Precision of these measurements was notably good, as illustrated in Fig. D-1, where deviations from the average of the duplicate determinations are shown for the 135 fuel-bearing segments from the LWBR fuel rods. When pooled over all 135 pairs of duplicates from all rods, the % RSD for a single determination of the UBT uranium concentration is only 0.045%. Considering that this standard deviation includes contributions from sampling errors, aliquot weighings, spike preparations, sample preparation chemistry (much of it in the hot-cell environment), and mass spectrometry, its small magnitude is particularly satisfying.

Systematic variances were associated with $\sigma_R^2(^{993}W_{SP})$, $\sigma_R^2(^{DS}R_{3/5})$, and $\sigma_R^2(^{segf}_{233})$. The systematic components of each of these variances were evaluated separately, as follows.

- a. Systematic Variance in the NBS-993 Spike Weight,
 $\sigma_{R,SYS}^2(^{993}W_{SP})$

Uncertainties in the NBS-993 spike weights were discussed in Section B.1.a. of this appendix. There, we indicated that uncertainties in the measurements by which the NBS-993 spikes were standardized to NBS 950a introduced a systematic variance into $^{993}W_{SP}$. The magnitude of this systematic variance was estimated from the standard deviation of the mean of the four measurements made in calibrating each batch of NBS-993 spikes. From the

Fig. D-1. Deviations from Average for Duplicate Determinations of UBT Uranium Concentration for LWBR Fuel Rod Segments

data in Table D-1, this standard deviation of the mean is calculated to be $0.019\%/ \sqrt{4}$, or 0.0095% , relative. Thus, in calculating our estimates of $\sigma_R^2([U]_{DS})$, we included a term corresponding to

$$\sigma_{R, SYS}^2(^{993}W_{SP}) = (0.0095\%)^2 \quad (D-29)$$

- b. Systematic Variance in the Measured Ratio of ^{233}U to ^{235}U in the DS Aliquot, $\sigma_{R, SYS}^2(^{DS}R_{3/5})$

The variance of uranium isotope ratios measured by TIMS was discussed in great detail in Appendix B. From that discussion, variance in the ratio $^{DS}R_{3/5}$ can be identified with the sum of (1) a systematic variance due to uncertainties in the fractionation correction applied to the measured ratio, (2) a systematic variance due to the isotopic procedure blank, and (3) a random variance that contains contributions from all random error sources, identified or not. The systematic variance $\sigma_{SYS}^2(^{DS}R_{3/5})$ may be expressed as

$$\sigma_{SYS}^2(^{DS}R_{3/5}) = \sigma_{SYS, MS}^2(^{DS}R_{3/5}) + \sigma_{SYS, Blank}^2(^{DS}R_{3/5}) \quad (D-30)$$

Equation B-9 in Appendix B was used to assign a value to $\sigma_{\text{SYS,MS}}^2(^{\text{DS}}\text{R}_{3/5})$. It is given by

$$\sigma_{\text{SYS,MS}}^2(^{\text{DS}}\text{R}_{3/5}) = (0.0000886)^2 \text{ } ^{\text{DS}}\text{R}_{3/5}^2 \quad (\text{D-31})$$

and, therefore, depends on the magnitude of the ratio. Similarly, the systematic variance due to the isotopic procedure blank is available from Eq. B-13 in Appendix B. In applying this expression to the present case, $^{\text{J}}\text{U}$ corresponds to ^{235}U and $^{\text{I}}\text{U}$ to ^{233}U , and $^{\text{J}}\text{U}_{\text{seg}}$ may be identified with the weight (nominally 1 mg) of ^{235}U in the NBS-993 spike. Substituting appropriate values into Eq. B-13, we have

$$\sigma_{\text{SYS,Blank}}^2(^{\text{DS}}\text{R}_{3/5}) = (6 \times 10^{-5})^2 + (6 \times 10^{-5} \text{ } ^{\text{DS}}\text{R}_{3/5})^2 \quad (\text{D-32})$$

Equation D-32 shows that the magnitude of this variance also depends on the value measured for $^{\text{DS}}\text{R}_{3/5}$.

Substituting Eqs. D-31 and D-32 into Eq. D-30, dividing both sides of the resulting equation by $^{\text{DS}}\text{R}_{3/5}^2$, and converting to % RSDs, we obtain an expression for the relative systematic variance of $^{\text{DS}}\text{R}_{3/5}$:

$$\sigma_{\text{R,SYS}}^2(^{\text{DS}}\text{R}_{3/5}) = \left(\frac{0.006\%}{^{\text{DS}}\text{R}_{3/5}} \right)^2 + (0.0107\%)^2 \quad (\text{D-33})$$

A term calculated from this expression, using the value of $^{\text{DS}}\text{R}_{3/5}$ appropriate to the segment in question, was also included in our estimate of $\sigma_{\text{R}}^2([U]_{\text{DS}})$ for each EOL segment.

c. Systematic Variance from the Weight Fraction of ^{233}U in the Segment Uranium, $\sigma_{\text{R,SYS}}^2(^{\text{segf}}_{233})$

In calculating $[U]_{\text{DS}}$ for any given segment, we used a value for $^{\text{segf}}_{233}$ for that was derived from WPC_{233} , as calculated for the specific segment under consideration. Variance in this value for $^{\text{segf}}_{233}$ also represented a systematic contribution to the variance in $[U]_{\text{DS}}$, since our replicate determinations of the uranium concentration did not include separate measurement of $^{\text{segf}}_{233}$ for each sample in a set. To account for this systematic variance, a term corresponding to $\sigma_{\text{R}}^2(\text{WPC}_{233})$, as evaluated for each segment according to Section B.2 of Appendix B, was incorporated into our estimated value of $\sigma_{\text{R}}^2([U]_{\text{DS}})$ as well.

Substituting all of the random and systematic variances introduced in the foregoing discussion into Eq. D-26 gives the expression we used in estimating $\sigma^2(U_{\text{SDS}})$:

$$\begin{aligned}
\sigma_R^2(U_{SDS}) &= \left(\frac{100\%}{W_{SDS}} \right)^2 [\sigma_{\text{weighing}}^2 + \sigma_{\text{Cal}}^2 + \sigma_{\text{Loss}}^2] \\
&+ (0.0095\%)^2 + (0.006\%/^{DS}R_{3/5})^2 + (0.0107\%)^2 \\
&+ \sigma_R^2(WPC_{233}) + \sigma_{R,Rdm}^2([U]_{UBT})
\end{aligned} \tag{D-34}$$

In applying this expression, the computer program selected values for the variables that corresponded to values for the specific segment to which a particular assay result applied. Thus, a different value for $\sigma_R^2(U_{SDS})$ was obtained for each segment. Typically, however, the term corresponding to $\sigma_R^2(W_{SDS})$ (see Eq. D-27) was $(2.1\%)^2$ or less, $^{DS}R_{3/5}$ was 0.001, $\sigma_R^2(WPC_{233})$ was $(0.03\%)^2$ or less (see Table B-3 in Appendix B), and $\sigma_{R,Rdm}^2([U]_{UBT})$ was $(0.045\%)^2$ (see Figure D-1). For a typical segment, then,

$$\begin{aligned}
\sigma_R^2(U_{SDS})|_{\max} &= (2.1\%)^2 + (0.0095\%)^2 + (6\%)^2 + (0.0107\%)^2 \\
&+ (0.03\%)^2 + (0.045\%)^2 = (6.36\%)^2
\end{aligned} \tag{D-35}$$

Clearly, most of the relative uncertainty in each value determined for U_{SDS} arose from potential solution losses during taking of the DS samples and from the isotopic procedure blank; the other variance terms are virtually negligible relative to these two. This result demonstrates the importance of our having included the systematic variances in our treatment of errors. Had the systematic variances been omitted, potential uncertainties in the assay results might have been grossly underestimated. We note at this point in our discussion that, although the relative uncertainty in U_{SDS} is substantial, it does not have a strong influence on the results for the total segment uranium assay because U_{SDS} accounts for only a tiny fraction of the total U, usually 0.1% or less. Thus, based on evaluation of the first term in Eq. D-25, uncertainties in U_{SDS} contributed a variance on the order of $(0.006\%)^2$ to the total variance in $^{235}U_{\text{seg},950}$.

2. Variance in U_{UBTS}

Just as the similarity between Eqs. D-10 and D-4 simplified our evaluation of bias in U_{UBTS} by permitting a comparison with the bias in U_{SDS} , it also simplifies evaluation of the variance in U_{UBTS} by allowing us to use many of the expressions derived in the foregoing section. In fact, the expression describing the variance in U_{UBTS} is completely analogous to that in Eq. D-34, with two exceptions.

The first exception stems from the use of ANL's internal-standard procedure for measuring the ratio $^{235}U/^{238}U$ by TIMS. In this procedure, fractionation corrections are applied on the basis of the observed ratio of isotopes in a standard isotopic mixture that is added to the mass spectrometer filament together with the unknown sample. It permits the fractionation correction factor for a given run to be determined from data generated within the same run. Since a separate correction factor is used for each TIMS analysis, the variance associated with this correction has no systematic-variance component and is fully accounted for in the random variance estimated from replicate measurements of

$[U]_{UBT}$. Thus, in estimating the variance for determination of U_{UBTS} , we set $\sigma_{SYS,MS}^2(UBT_{R_{3/5}})$ equal to zero (cf. Eq. D-31).

With regard to the second exception, duplicate determinations of $[U]_{UBT}$ were made for all segments. Hence, the random variance in $[U]_{UBT}$, where $[U]_{UBT}$ was calculated as the average of the two measured values, was computed as $\sigma_{R,Rdm}^2([U]_{UBT})/2$.

Taking these differences into account provides the following expression for the relative variance in U_{UBTS} :

$$\begin{aligned}\sigma_R^2(U_{UBTS}) = & \left(\frac{100\%}{W_{UBTS}} \right)^2 [\sigma_{Weighing}^2 + \sigma_{Cal}^2 + \sigma_{Loss}^2] + (0.0095\%)^2 \\ & + (0.006\%/UBT_{R_{3/5}})^2 + (0.006\%)^2 + \sigma_R^2(WPC_{233}) \\ & + \sigma_{R,Rdm}^2([U]_{UBT})/2\end{aligned}\quad (D-36)$$

Again, because our computer program assigned values appropriate to each particular segment in applying this expression to estimating $\sigma_R^2(U_{UBTS})$, a separate variance estimate was obtained for each segment. Typically, for the U_{UBTS} variance estimates, the term corresponding to $\sigma_R^2(W_{UBTS})$ (see Eq. D-27) was $(1.1\%)^2$ or less as stated in Section V.C.3.a.(2) of the report body; $UBT_{R_{3/5}}$ was approximately unity; $\sigma_R^2(WPC_{233})$ was $(0.03\%)^2$ or less (see Table B-3 in Appendix B); and $\sigma_R^2([U]_{UBT})$ was 0.045% (see Fig. D-1). For such a segment, one obtains

$$\begin{aligned}\sigma_R^2(U_{UBTS})|_{\max} = & (1.1\%)^2 + (0.0095\%)^2 + (0.006\%)^2 + (0.006\%)^2 \\ & + (0.03\%)^2 + (0.0045\%)^2/2 = (1.1\%)\end{aligned}\quad (D-37)$$

Uncertainty in the value determined for U_{UBTS} was, thus, almost entirely due to potential solution losses during sampling of the UBT solution. Since U_{UBTS} corresponded to approximately 2 to 3% of the total segment uranium as determined by using the 950a-MSID assay method, the 1.1% RSD estimated in Eq. D-37 contributes a variance in the range $(0.02\%)^2$ to $(0.03\%)^2$ to $\sigma_R^2(^T U_{seg,950})$ according to Eq. D-25. This contribution accounts for a substantial fraction of the total variance in $^T U_{seg,950}$; in some cases, it accounts for the predominant portion of the uncertainty in the 950a-MSID assay result.

3. Variance in U_{SBT}

On the basis of the relationship in Eq. D-12 and the same approximations that were employed in deriving Eq. D-13, the relative variance of U_{SBT} may be described by the relationship

$$\sigma_R^2(U_{SBT}) = \sigma_R^2(^{950}W_{SP}) + \sigma_R^2(^{seg}f_{233}) + \sigma_R^2(^{SBT}R_{3/8})\quad (D-38)$$

Variance in the weight of the NBS-950a spike, $\sigma_R^2(^{950}\text{W}_{\text{SP}})$, arose primarily from random errors associated with weighing the $^{238}\text{U}_3\text{O}_8$ present in the capsule that contained the spike. This weighing (performed using a five-place semi-micro balance) showed an estimated standard deviation of 0.00005 g. For spikes that contained a nominal 1-g quantity of uranium (1.2 g U_3O_8), this standard deviation translates to a % RSD in the spike weight of 0.0042%; for 3-g spikes (3.5 g U_3O_8), the corresponding % RSD is 0.0014%. In calculating its estimate of $\sigma_R^2(\text{U}_{\text{SBT}})$, our computer program assigned a systematic variance term corresponding to $\sigma_R^2(^{950}\text{W}_{\text{SP}})$, as appropriate to the spike size for the particular segment under consideration.

A second systematic variance term was also included to account for the contribution of $\sigma_R^2(^{\text{segf}}\text{f}_{233})$. As in previously discussed calculations that called for values of this variance, the program used the estimated value of $\sigma_R^2(\text{WPC}_{233})$ for the specific segment under consideration in assigning the value of this term.

Like $\text{UBT}\text{R}_{3/5}$, which was treated in the previous section, $\text{SBT}\text{R}_{3/8}$ was measured using the ANL internal-standard procedure for the TIMS ratio measurements and was determined for each of two completely separate samples of solution from the blend tank. By analogy with the treatment of $\text{UBT}\text{R}_{3/5}$, the variance in $\text{SBT}\text{R}_{3/8}$, was treated as consisting of a systematic variance arising from the isotopic procedure blank, as evaluated from the appropriate form of Eq. B-13 in Appendix B, and a random variance that includes contributions from all random error sources that affect the observed differences between replicate determinations of $\text{SBT}\text{R}_{3/8}$, such as homogeneity of the SBT solution, TIMS measurements, etc. In applying Eq. B-13 to calculating the systematic variance in $\text{SBT}\text{R}_{3/8}$, we let $^{\text{J}}\text{U}$ correspond to ^{238}U , $^{\text{I}}\text{U}$ to ^{233}U , and $^{\text{J}}\text{U}_{\text{seg}}$ to the weight of ^{238}U in the aliquot of SBT solution that was taken for analysis, which we designate $^{238}\text{U}_{\text{SBT,ALQ}}$. This weight of ^{238}U varied from aliquot to aliquot, depending upon the weight of solution dispensed from the SBT solution sample during aliquoting and on the concentration of ^{238}U in the SBT. Substituting appropriate variables into Eq. B-13, assuming that blank levels of ^{233}U and ^{238}U in each aliquot corresponded to 6×10^{-8} g U as discussed in Section B.2 of Appendix B, dividing both sides of the resulting equation by $\text{SBT}\text{R}_{3/8}$, and converting to % RSDs, we obtain the expression

$$\sigma_{R,\text{SYS}}^2(\text{SBT}\text{R}_{3/8}) = \frac{1}{^{238}\text{U}_{\text{SBT,ALQ}}^2} \left[\left(\frac{6 \times 10^{-6}\%}{\text{SBT}\text{R}_{3/8}} \right)^2 + (6 \times 10^{-6} \%)^2 \right] \quad (\text{D-39})$$

In evaluating this systematic variance term for inclusion in our estimate of the uncertainty in U_{SBT} , our computer program used the average value of $\text{SBT}\text{R}_{3/8}$ obtained from replicate measurements of this ratio for the particular segment under consideration. It evaluated $^{238}\text{U}_{\text{SBT,ALQ}}$ by first computing the concentration of ^{238}U in the SBT solution as the weight of ^{238}U in the 950a spike divided by the weight of solution in the SBT (this value was recorded as part of the DDS operations) and then multiplying this concentration by the RMS weight of solution taken for the replicate aliquots appropriate to the given segment.

Combining all the systematic and random variances introduced in the foregoing discussion according to Eq. D-38 and taking into account the duplicate determinations that were made of $^{SBTR}_{3/8}$, we obtain the expression

$$\sigma_R^2(U_{SBT}) = \sigma_R^2(^{950}W_{SP}) + \sigma_R^2(^{segf}_{233}) + \left(\frac{W_{SBT}}{W_{ALQ,RMS} \ ^{950}W_{SP}} \right)^2 \left[\left(\frac{6 \times 10^{-6}\%}{^{SBTR}_{3/8}} \right)^2 + (6 \times 10^{-6}\%)^2 \right] + \sigma_{R,Rdm}^2 (^{SBTR}_{3/8})/2 \quad (D-40)$$

Here, we expanded the term in front of the brackets in Eq. D-39 to better reflect the way in which $^{238}U_{SBT,ALQ}^2$ was calculated in our computer program: W_{SBT} represents the weight of solution in the SBT as recorded during DDS operations; $W_{ALQ,RMS}$ represents the RMS weight of SBT solution aliquots processed for the specific segment being considered; and $^{950}W_{SP}$ stands for the weight of uranium (presumed for this calculation to be all ^{238}U) in the NBS-950a spike added to the blend tank.

For a typical segment, $\sigma_R^2(^{950}W_{SP})$ had the value $(0.0042\%)^2$ or $(0.0014\%)^2$, depending on whether a 1-g or 3-g spike was used (see above); $\sigma_R^2(^{segf}_{233})$ was 0.03% or less as indicated in Table B-3 of Appendix B; $W_{ALQ,RMS}$ was chosen to provide a value for $U_{SBT,ALQ}$ of 1 mg; $^{950}W_{SP}$ was 1 g or 3 g; and $^{SBTR}_{3/8}$ fell in the range 0.2 to 5.

The random variance, $\sigma_{R,Rdm}^2(^{SBTR}_{3/8})$ was estimated from the pooled variance, over segments from a given rod, of corresponding ratios measured for duplicate samples from the SBT for each segment. The exceptional precision that we obtained by using the internal-standard procedure for these TIMS ratio measurements is demonstrated in Fig. D-2, which shows deviations from the average of the duplicate ratios for the 135 fuel-bearing segments from the LWBR fuel rods. When pooled over all 135 pairs of duplicates from all rods, the % RSD for a single determination of $^{SBTR}_{3/8}$ is only 0.0062%.

Substituting these typical values into Eq. D-40 and choosing values to maximize the magnitude of $\sigma_R^2(U_{SBT})$ give the result

$$\begin{aligned} \sigma_R(U_{SBT})|_{\max} &= (0.0042\%)^2 + (0.03\%)^2 \\ &+ \frac{1}{(0.001)^2} \left[\frac{(6 \times 10^{-6}\%)^2}{(0.2)^2} + (6 \times 10^{-6}\%)^2 \right] + (0.0062\%)^2/2 \\ &= (0.043\%)^2 \end{aligned} \quad (D-41)$$

Uncertainty in the value determined for U_{SBT} was largely due to the systematic variances associated with the MSID method. Random errors made only a small contribution to the total variance, as indicated by the relative magnitudes of

Fig. D-2. Deviations from Average for $^{233}\text{U}/^{238}\text{U}$ Ratios Determined Using Duplicate Samples of Spiked Blend Tank Solution

the terms in Eq. D-41. Because U_{SBT} accounted for the bulk (97 to 98%) of the total segment uranium, its variance contributed strongly to the variance in $^{233}\text{U}_{\text{seg},950}$, as shown in the next section.

4. Combination of Variances Contributing to $\sigma_R^2(^{233}\text{U}_{\text{seg},950})$

The uncertainty we quoted for the 950a-MSID assay result from each segment was computed by combining all the individual variance contributions discussed above according to Eq. D-25. A separate standard deviation was estimated for each segment assay because of the appearance of segment-dependent variables in most of the expressions involved in the calculations. Use of the project computer system in carrying out the calculations permitted convenient, error-free access to the requisite segment-specific data and ensured that each uncertainty estimated was both realistic and reliable.

Using information from Eqs. D-35, D-37, and D-41 together with typical segment variables defined in the foregoing discussion, the magnitude of $\sigma_R^2(^{233}\text{U}_{\text{seg},950})$ for a representative segment according to Eq. D-25 is

$$\begin{aligned} \sigma_R^2(^{233}\text{U}_{\text{seg},950})_{\text{max}} &= (0.001)^2 (6.36\%)^2 + (0.025)^2 (1.1\%)^2 \\ &\quad + (0.974)^2 (0.043\%)^2 = (0.051\%)^2 \end{aligned} \quad (\text{D-42})$$

This result suggests that the project requirement restricting the % RSD of the 950a-MSID result to 0.15% or less should easily have been met by the ANL assays. This expectation is borne out by the data in Table D-5, which shows the % RSD estimated for the 950a-MSID assay result for each fuel-bearing segment processed during the EOL campaign. Most of the values shown in this table are consistent with the representative value calculated in Eq. D-42.

For a few segments of low uranium content the estimated standard deviation is larger than this representative value. The increased standard deviation in these cases resulted from the low concentration of segment uranium in the SBT solution, which precluded our using an aliquot containing an amount of uranium sufficient to "swamp out" the effects of the isotopic procedure blank. Aliquot sizes were recommended to the analyst preparing samples for the assay measurements on the basis of approximate segment-uranium loading values provided by BAPL. Typically, the aliquots were sized to contain 1 mg of segment uranium but, for practical handling, were restricted to the range between 1 and 10 g of solution. For only one (C-01) of the 135 fuel-bearing segments processed during the EOL campaign was the uranium content of the aliquot so small as to cause the limit (0.15%, relative) to allowable standard deviation for the segment uranium assay to be exceeded. Because the uranium content of this segment (0.22 g) accounted for only a minor fraction of the uranium in the whole rod (58.22 g), the larger relative standard deviation (0.185%) associated with it was judged to be acceptable.

For the eight fuel-bearing segments that were involved in dissolver-operation mishaps, additional variance terms were included in estimating the quoted standard deviations to account for uncertainties in any additional measurements that were performed and in our estimation of potentially unrecovered fuel material (see Section V.C.3.b of the report body). Evaluation of each of these additional terms was described in detail in the individual rod reports; the standard deviations in Table D-5 for these segments include contributions from these terms. We note that the estimated standard deviations for all of the affected segments are smaller than the 0.15% limit to allowable error.

All in all, we hope to have shown in the foregoing discussion that the standard deviations quoted for the 950a-MSID assay results reported by ANL during the EOL campaign were legitimately derived and properly calculated, and that they included contributions from all significant sources of random and randomly varying systematic error. To support the validity of these estimates, we call attention to the results of assays performed during the qualification and mid-campaign verification of the ANL equipment and procedures. These results are shown in Table D-6. The segments listed in this table were prepared at ANL from non-irradiated thoria and binary ($\text{ThO}_2 \cdot ^{233}\text{UO}_2$) pellets provided by and previously characterized at BAPL. Each of these segments was dissolved and analyzed using the same procedures and equipment that were employed for the EOL LWBR fuel rod segments. Segments U-01, U-03, V-05, and V-06 were processed during qualification of these systems in the fall of 1983; W-01 and W-04 were processed in the mid-campaign verification during June 1985. (Segments W-02 and W-03 of the performance verification were similarly prepared and processed but contained pellets from a batch different from the pellets in the listed segments; comparative data for this second batch of pellets included only these two segments and are considered insufficient to reliably estimate the

Table D-5. Estimated Standard Deviations of the 950a-MSID Uranium Assay Results for Individual Fuel Rod Segments

Rod No.	Estimated RSD for Indicated Segment, %														
	01	02	03	04	05	06	07	08	09	10	11	12	13	14	15
B	0.043	0.028	0.028	0.027	0.031	0.027	0.030	—	—	—	—	—	—	—	—
C	0.185 ^a	0.026	0.030 ^b	0.028 ^b	0.027	0.028	0.029	—	—	—	—	—	—	—	—
D	0.134	0.026	0.028	0.027	0.028	0.027	0.050	—	—	—	—	—	—	—	—
E	0.038	0.026	0.026	0.027	0.023	0.023	0.027	—	—	—	—	—	—	—	—
F	0.051	0.029	0.030	0.031	0.030	0.027	0.030	0.029	—	—	—	—	—	—	—
G	0.036	0.025	0.025	0.024	0.026	0.024	0.021	—	—	—	—	—	—	—	—
H	0.055	0.023	0.021	0.022	0.018	0.019	0.020	0.086 ^b	0.020	0.019	0.020	0.020	0.024	0.020	0.037
I	0.029	0.025	0.025	0.025	0.026	0.025	0.023	0.023	—	—	—	—	—	—	—
J	0.026	0.028	0.073 ^b	0.029	0.028	0.024	0.020	0.034 ^b	—	—	—	—	—	—	—
K	0.029	0.026	0.027	0.024	0.026	0.026	0.030	0.028	—	—	—	—	—	—	—
L	0.036	0.030	0.029	0.030	0.030	0.025	0.028	0.026	—	—	—	—	—	—	—
M	0.030	0.024	0.026	0.026	0.024	0.036	—	—	—	—	—	—	—	—	—
N	0.028	0.022 ^b	0.023	0.023	0.024	0.052 ^b	—	—	—	—	—	—	—	—	—
O	0.024	0.030	0.132 ^b	0.025	0.027	0.039	—	—	—	—	—	—	—	—	—
P	0.019	0.024	0.022	0.022	0.023	0.031	—	—	—	—	—	—	—	—	—
Q	0.026	0.030	0.030	0.029	0.029	0.043	—	—	—	—	—	—	—	—	—
R	0.146	0.025	0.022	0.023	0.025	0.021	0.021	0.022	0.022	0.021	0.022	0.025	0.024	0.022	0.046
W ^c	0.026	0.064	0.027	0.027	—	—	—	—	—	—	—	—	—	—	—

^aValue exceeds limit (0.15%) to allowable standard deviation for uranium assay.

^bValue includes contributions from recovery operations that followed dissolution mishap.

^cNonirradiated test rod used in mid-campaign performance verification.

Table D-6. Results of 950a-MSID Uranium Assays on Segments Prepared from Nonirradiated Binary Fuel Pellets

Segment	Nominal U Loading in Segment, ^a g	Total Segment U by 950a-MSID, ^b g	% Difference (Found - Nominal)
U-01 ^c	2.39196	2.39537	+0.143
U-03 ^c	2.37317	2.37712	+0.166
V-05 ^c	2.39707	2.40174	+0.195
V-06 ^c	2.39869	2.40235	+0.153
W-01 ^d	3.19901	3.20487	+0.183
W-04 ^d	3.19625	3.20115	+0.153
Average Difference:			+0.166%
Std. Dev. of Diff:			+0.020%

^aCalculated based on BAPL estimate of U content for blend from which pellets were taken; confidence interval extends $\pm 0.23\%$ from nominal value.

^bCorrected for fuel losses during shearing when appropriate.

^cProcessed during qualification of ANL equipment and procedures.

^dProcessed during mid-campaign performance verification.

standard deviation of the measurements.) The observed standard deviation of the 950a-MSID results from these separately processed but similar segments is 0.020%, relative, and is remarkably consistent with the standard deviations (0.027%, relative) estimated for the Rod W segments by our computer program (see Table D-5). This consistency lends strong credibility to our computer-generated standard deviation estimates and strengthens our conclusion that the project requirements on precision of the segment uranium assay were met.

F. Standard Deviation of the 993-MSID Assay Results

The standard deviations of uranium results determined for each segment by the 993-MSID assay method were also estimated by our computer program. The expressions used in calculating these estimates were derived as follows.

When the weight of uranium in a dissolved segment is calculated from Eq. D-16, the relative variance in the result may be written in terms of the relative variances of the contributing variables as

$$\sigma_R^2(T_{U_{\text{seg},993}}) = \left(\frac{U_{\text{SDS}}}{T_{U_{\text{seg},993}}} \right)^2 \sigma_R^2(U_{\text{SDS}}) + \left(\frac{U_{\text{UBT}}}{T_{U_{\text{seg},993}}} \right)^2 [\sigma_R^2(W_{\text{UBT}}) + \sigma_R^2([U]_{\text{UBT}})] \quad (\text{D-43})$$

Each of the terms in this expression has been discussed earlier except for the variance, $\sigma_R^2(W_{\text{UBT}})$, associated with the weight of solution present in the UBT

prior to its having been sampled. In pre-EOL characterization of the blend-tank weighing systems, we found that this variance corresponded to several grams in the UBT solution weight, even though the balance used for the weighing had a reproducibility of less than 1 g. We were not able to pinpoint the reason for this imprecision, although we found some indications that it may have been related to changes in the moisture content of insulating materials in the blend-tank heaters. In any case, for the EOL estimates, we assigned a standard deviation of 7 g to the UBT solution weighing and computed the relative variance in W_{UBT} from the relationship

$$\sigma_R^2(W_{\text{UBT}}) = \left(\frac{7 \times 100\%}{W_{\text{UBT}}} \right)^2 \quad (\text{D-44})$$

For typical UBT solution weights between 5 and 8 kg, this variance corresponded to a standard deviation of roughly 0.1%, relative.

Using the expression for $\sigma_R^2(W_{\text{UBT}})$ in Eq. D-44, the expression for $\sigma_R^2(U_{\text{SDS}})$ in Eq. D-34, and the expression for $\sigma_R^2([U]_{\text{UBT}})$ given by the last five terms of Eq. D-36, our computer program calculated a separate estimate of $\sigma_R^2(TU_{\text{seg},993})$ for each segment. As the reader can easily verify, the terms related to U_{SDS} in Eq. D-43 made a negligible contribution to this estimated variance in every case because of the small magnitude of the factor given by $(U_{\text{SDS}}/TU_{\text{seg},993})^2$. To a good approximation, then, the estimated value of $\sigma_R^2(TU_{\text{seg},993})$ was given by the expression

$$\begin{aligned} \sigma_R^2(TU_{\text{seg},993}) = & \left(\frac{7 \times 100\%}{W_{\text{UBT}}} \right)^2 + (0.0095\%)^2 + (0.006\%/^{\text{UBT}}R_{3/5})^2 + (0.006\%)^2 \\ & + \sigma_R^2(WPC_{233}) + \sigma_{R,\text{Rdm}}^2([U]_{\text{UBT}})^2/2 \end{aligned} \quad (\text{D-45})$$

Substituting typical values for the individual variables involved in this expression, one has

$$\begin{aligned} \sigma_R^2(TU_{\text{seg},993}) \simeq & (0.1\%)^2 + (0.0095\%)^2 + (0.006\%)^2 + (0.006\%)^2 \\ & + (0.03\%)^2 + (0.045\%)^2/2 \simeq (0.11\%)^2 \end{aligned} \quad (\text{D-46})$$

This illustrative evaluation of Eq. D-43 suggests that the primary source of imprecision in the 993-MSID assay results was imprecision of the UBT solution weighing. The magnitude of the variance associated with this weighing, moreover, could not be estimated for any given segment during the campaign operations because, over the duration of the weighing operations, it represented a systematic error in the measured weights. The standard deviations that we quoted during the campaign for the 993-MSID uranium assay results were, therefore, only roughly estimated. The question of how good these estimates may have been is addressed in the following section, where the 993-MSID assay results obtained during the campaign are compared with the 950a-MSID results.

G. Comparison of 993-MSID and 950a-MSID Uranium Assay Results

Before we present the comparisons of actual 993-MSID and 950a-MSID results obtained for the uranium assays of LWBR fuel rod segments, let us first consider the differences that might be expected between the methods on the basis of the error analysis that was presented in the foregoing discussions.

A limit to the magnitude of the average observed difference between results from the two methods may be obtained from the potential biases associated with the results. The expected magnitude of the percent relative difference, D , between two results of closely similar value is given by the difference in the percent relative biases of the individual results. Thus, we may write

$$D_{(993-950)} = B_R(^T U_{\text{seg},993}) - B_R(^T U_{\text{seg},950}) \quad (\text{D-47})$$

From Eqs. D-17, D-18, and D-7,

$$\begin{aligned} B_R(^T U_{\text{seg},993}) \simeq & B_R(W_{\text{UBT}}) + B_R([U]_{\text{UBT}}) = B_R(W_{\text{UBT}}) + B_R(^{993}W_{\text{SP}}) \\ & + B_R(^{993}f_{235}) + B_R(^{\text{UBT}}R_{3/5}) - B_R(^{\text{UBT}}W_{\text{ALQ}}) - B_R(^{\text{seg}}f_{233}) \end{aligned} \quad (\text{D-48})$$

From Eq. D-20,

$$B_R(^T U_{\text{seg},950}) \simeq B_R(^{950}W_{\text{SP}}) + B_R(^{950}f_{238}) + B_R(^{\text{SBT}}R_{3/8}) - B_R(^{\text{seg}}f_{233}) \quad (\text{D-49})$$

Substituting these expressions into Eq. D-47 and simplifying by noting that $B_R(^{993}W_{\text{SP}})$ and $B_R(^{950}W_{\text{SP}})$ are identical owing to our having cross-standardized the NBS-993 and NBS-950a reference materials for this work, we may express the difference $D_{(993-950)}$ as

$$\begin{aligned} D_{(993-950)} = & B_R(W_{\text{UBT}}) + B_R(^{993}f_{235}) + B_R(^{\text{UBT}}R_{3/5}) - B_R(^{\text{UBT}}W_{\text{ALQ}}) \\ & - B_R(^{950}f_{238}) - B_R(^{\text{SBT}}R_{3/8}) \end{aligned} \quad (\text{D-50})$$

Each of these bias terms that contribute to an expected difference between the methods has been evaluated in our earlier treatments. Noting that $B_R(^{\text{UBT}}R_{3/5})$ and $B_R(^{\text{SBT}}R_{3/8})$ are related according to Eq. B-5 of Appendix B and always have the same sign, we may substitute values for the individual terms to obtain an estimate of the maximum probable difference that should be observed between results from the two methods. Choosing signs for the various contributions to maximize the value of $D_{(993-950)}$, we have

$$\begin{aligned} D_{(993-950)}|_{\text{max}} = & 0.02\% + 0.001\% + (-2/3)(0.05\%) - 0\% + 0.001\% \\ & - (-5/3)(0.05\%) = 0.072\% \end{aligned} \quad (\text{D-51})$$

Thus, on the average, we might expect a percent relative difference between the 993-MSID and 950a-MSID assay results as large as 0.072% merely from uncertainties in the standards and bias errors in the measurements involved in applying the methods.

The percent relative differences between the 993-MSID and 950a-MSID assay results for each of the 135 fuel-bearing LWBR fuel rod segments processed at ANL are shown graphically in Fig. D-3. In this figure, two values are shown for each segment to illustrate the magnitudes of variations between 993-MSID results that were observed if one or the other of the two $[U]_{UBT}$ values measured for each segment (cf. Fig. D-1) were used in calculating $^{235}U_{seg,993}$. Because of the high precision of the TIMS uranium isotope ratio measurements involved in determining U_{SPT} for the 950a-MSID assays (cf. Fig. D-2), similar variations in the values for $^{235}U_{seg,950}$ were inconsequential.

Fig. D-3. Deviations Between 950a-MSID and 993-MSID Uranium Assay Results for LWBR Fuel Rod Segments

The average relative difference (relative to the 950a-MSID assay result) over these 135 segments was 0.0175%, with the 950a-MSID results being the larger. This difference is considerably smaller than the maximum probable bias (0.072%) between these methods, as estimated in Eq. D-51, and provides no evidence for significant bias errors, in either method, other than those that were accounted for in our error analysis. Thus, corroboration of the 950a-MSID uranium assay results by the 993-MSID method is totally satisfactory.

Variations in the observed $TU_{seg,993} - TU_{seg,950}$ differences among the individual segments may be related to the variance of the two assay results according to

$$\sigma^2(D_{(993-950)}) = \sigma_R^2(TU_{seg,993}) + \sigma_R^2(TU_{seg,950}) = (0.0889\%)^2 \quad (D-52)$$

where the standard deviation of the relative difference was evaluated from the data in Fig. D-3, ignoring the non-zero average difference between methods; if this difference is taken into account, the standard deviation estimated from these data decreases slightly, to 0.0875%. Noting that $\sigma_R^2(TU_{seg,993})$ is very nearly identical to $\sigma_R^2(W_{UBT})$ (cf. Eq. D-46), we may use this relationship together with the data regarding $\sigma_R^2(TU_{seg,950})$ in Table D-5 to derive an estimate of $\sigma_R^2(W_{UBT})$. To account for the different magnitudes that $\sigma_R^2(TU_{seg,950})$ has for the individual segments, we write Eq. D-52 in terms of the sums of variances over all the segments under consideration

$$\sum_{seg} \sigma^2(D_{(993-950)}) = \sum_{seg} \sigma_R^2(TU_{seg,993}) + \sum_{seg} \sigma_R^2(TU_{seg,950}) \quad (D-53)$$

Then, assuming that $\sigma^2(D_{(993-950)})$ and $\sigma_R^2(TU_{seg,993})$ are constant for all segments and substituting $\sigma_R^2(W_{UBT})$ for $\sigma_R^2(TU_{seg,993})$, we have

$$N_{seg} \sigma^2(D_{(993-950)}) = N_{seg} \sigma_R^2(W_{UBT}) + \sum_{seg} \sigma_R^2(TU_{seg,950}) \quad (D-54)$$

where N_{seg} is the number of segments in the summation (135 in the present case), and the individual $\sigma_R^2(TU_{seg,950})$ values are given in Table D-5. Solving Eq. D-54 for $\sigma_R^2(W_{UBT})$ gives

$$\sigma_R^2(W_{UBT}) = \sigma^2(D_{(993-950)}) - [\sum_{seg} \sigma_R^2(TU_{seg,950})]/N_{seg} \quad (D-55)$$

Substituting values for the variables in this expression, we get

$$\sigma_R^2(W_{UBT}) = (0.0889\%)^2 - (0.0390\%)^2 = (0.0799\%)^2 \quad (D-56)$$

The value 0.0799% for $\sigma_R(W_{UBT})$ obtained in this way from our EOL data is in fair agreement with the values (roughly 0.1%) that were assumed in our computerized uncertainty calculations made during the campaign (see Eq. D-44). This result suggests that the uncertainties we assigned to the 993-MSID assay results were reasonably good estimates of the corresponding standard deviations. Overall, agreement between the 950a-MSID and 993-MSID results for the fuel-bearing segments processed during the EOL campaign is quite consistent with expectations based on the error analysis presented in this Appendix. This consistency lends strong support to the validity of the treatment we applied.

APPENDIX E

ESTIMATION OF BIAS AND STANDARD DEVIATION FOR FISSION PRODUCT DETERMINATIONS

A. Introduction

For each fuel-bearing segment that was processed during the EOL campaign, ANL determined the number of atoms of each of three fission products (^{137}Cs , ^{144}Ce , and ^{95}Zr) present in the segment. As with our uranium assays and isotopic measurements, the results for each fission product were calculated using a computer program that not only compiled the necessary data and performed data manipulations appropriate to providing the individual fission product loadings, but also estimated the standard deviation of the calculated results, including both random and randomly varying systematic error contributions in the estimate. In this appendix, we derive the expressions that were used by this report-generator computer program in evaluating the quoted standard deviations and report estimates of the potential bias associated with each fission-product assay result.

Because the same general approach was used for computing the loadings of all three fission products in each segment, we begin our treatment by deriving general expressions for the bias and standard deviation. Then, we expand the discussion to accommodate specific details of the way in which individual terms in the general expressions were evaluated for a particular nuclide. The resulting estimates of maximum probable bias and standard deviation are shown to meet pertinent project error requirements in all cases.

B. General Equations for Bias and Standard Deviation of Fission Product Determinations

For each segment, the total number of atoms of a given fission product was calculated according to the relationship

$$FP_{\text{seg}} = [FP]_{\text{UBT,seg}} F_{\text{FP,Rod}} W_{\text{UBT,seg}} (1 + U_{\text{SDS}}/U_{\text{UBT}})_{\text{seg}} \quad (\text{E-1})$$

In this expression, FP_{seg} stands for the atoms of particular fission product in the segment of interest; $[FP]_{\text{UBT,seg}}$ is the apparent (i.e., measured) concentration (atoms per gram of solution) of this fission product in the UBT solution from the segment's dissolution; $F_{\text{FP,Rod}}$ is a correction factor to account for apparent bias error in the calibration of the gamma spectrometer used for measuring $[FP]_{\text{UBT,seg}}$ and was separately evaluated for measurements related to each individual fuel rod; $W_{\text{UBT,seg}}$ is the weight of solution present in the UBT before sampling; and the factor $(1 + U_{\text{SDS}}/U_{\text{UBT}})$ represents a correction for the fraction of segment fuel that was removed from the dissolver system in sampling the second dissolution, as evaluated from uranium assay data on the DS samples and on the UBT solution. In this final term of Eq. E-1, U_{SDS} stands for the amount of uranium present in the DS samples, and U_{UBT} represents the uranium in the unsampled UBT solution (see Section B.1 and B.2 of Appendix D for discussion of these uranium assay values and details of their determination).

From a Taylor-series expansion of Eq. E-1, errors in FP_{seg} may be expressed in terms of the errors associated with the individual factors that form the product in the right side of this equation. When derived in this manner, bias in FP_{seg} is given by

$$B_R(FP_{seg}) = B_R([FP]_{UBT,seg}) + B_R(F_{FP,Rod}) + B_R(W_{UBT,seg}) \\ + B_R[(1 + U_{SDS}/U_{UBT})_{seg}] \quad (E-2)$$

Here, we use the notation $B_R(X)$ to represent the percent relative bias in the variable X , where the variables are defined as in Eq. E-1. Similarly, the percent relative standard deviation, represented by $\sigma_R(X)$, for FP_{seg} can be related to the corresponding values for the individual factors through

$$\sigma_R^2(FP_{seg}) = \sigma_R^2([FP]_{UBT,seg}) + \sigma_R^2(F_{FP,Rod}) + \sigma_R^2(W_{UBT,seg}) \\ + \sigma_R^2[(1 + U_{SDS}/U_{UBT})_{seg}] \quad (E-3)$$

The last two terms in each of these expressions, i.e., the terms related to W_{UBT} and to $(1 + U_{SDS}/U_{UBT})$, are independent of the identity of the fission product under consideration and, thus, have a set value for all fission products from a particular segment. The factor $(1 + U_{SDS}/U_{UBT})$ represented only a very minor correction to the fission product loadings, increasing the value of FP_{seg} by only 0.1% or less in most cases. Thus, errors associated with this correction were deemed negligible relative to the other terms in Eqs. E-2 and E-3 and are ignored in the remainder of our current discussion.

Errors in the UBT solution weight, $W_{UBT,seg}$, were discussed in Sections C (bias) and F (standard deviation) of Appendix D. In those discussions, $B_R(W_{UBT,seg})$ was assigned the value 0.02%, corresponding to the potential bias associated with the weighing of the UBT solution, and $\sigma_R^2(W_{UBT,seg})$ was estimated on the basis of the relationship

$$\sigma_R^2(W_{UBT,seg}) = \left(\frac{7 \times 100\%}{W_{UBT,seg}} \right)^2 \quad (E-4)$$

This relationship expresses the dependence of $\sigma_R^2(W_{UBT,seg})$ on the value of $W_{UBT,seg}$ for the specific segment under consideration; we employed it in our computer program to obtain a separate value for $\sigma_R^2(W_{UBT,seg})$ for each segment we processed.

The factors $[FP]_{UBT,seg}$ and $F_{FP,Rod}$ in Eq. E-1 were not measured directly but were calculated from the results of measurements on weighed aliquots of the UBT solution samples and on working standards that contained known quantities of the fission products of interest. Evaluation of the bias and variance terms related to these factors for use in applying Eqs. E-2 and E-3 is best carried

out by propagating the errors associated with these measurements to the calculated factors. We consider the errors in each of these factors separately in Sections C and D below and combine them with errors associated with $W_{\text{UBT,seg}}$ in Section E to provide expressions for $B_R(\text{FP}_{\text{seg}})$ and $\sigma_R^2(\text{FP}_{\text{seg}})$.

C. Errors in the Measured Fission Product Concentrations, $[\text{FP}]_{\text{UBT,seg}}$

The concentration of each fission product in the UBT solution was determined by gamma counting weighed aliquots of the UBT solution samples as described in Section IV.F.1.e of the report body. One aliquot from each of two separate UBT samples was counted to determine ^{137}Cs and ^{144}Ce , which could be measured simultaneously with no chemical pretreatment of the aliquot other than diluting it to a predetermined volume. Using a hydroxide precipitation procedure, we prepared a separate aliquot from each of the same two samples for the ^{95}Zr determination by removing the bulk of its cesium activity (^{134}Cs and ^{137}Cs). This treatment of these aliquots reduced the total count rate obtained for them (thereby permitting larger aliquots to be used) and lowered the Compton background under the ^{95}Zr gamma peaks in their spectra; these effects helped improve the sensitivity of the ^{95}Zr measurements. Both ^{95}Zr and ^{144}Ce were quantitatively carried with the precipitate, along with several other fission products (e.g., ^{154}Eu) that were not of direct interest. The ^{144}Ce in each precipitated aliquot was used at a yield monitor to account for small (15% or less) losses that occurred during subsequent handling and redissolution of the precipitate.

The number, $N_{\text{ALQ,FP}}$, of atoms of each fission product (corrected for radioactive decay to January 1, 1984) in each aliquot was automatically computed from the gamma-ray spectrum of the aliquot by the GeLiGAM software associated with our gamma spectrometer system (see Section III.C.2.b.(2) of the report body for the software description). The GeLiGAM programs also estimated the counting error associated with each value of $N_{\text{ALQ,FP}}$, including error contributions related to corrections that the programs made for background and interfering peaks in the spectrum. The apparent concentration of each nuclide in a given aliquot, $[\text{FP}]_{\text{ALQ}}$, was computed by dividing $N_{\text{ALQ,FP}}$ by the aliquot weight, W_{ALQ} , which had been recorded when the aliquot was taken from the corresponding UBT solution sample. This expression is

$$[\text{FP}]_{\text{ALQ}} = N_{\text{ALQ,FP}}/W_{\text{ALQ}} \quad (\text{E-5})$$

For the precipitated aliquots, the GeLiGAM program subsequently corrected the apparent concentration of ^{95}Zr for losses that were incurred, during handling of the precipitate, by multiplying the right side of Eq. E-5 by the ratio of apparent ^{144}Ce concentrations in the untreated and precipitated aliquots. That is,

$$[^{95}\text{Zr}]_{\text{ALQ}} = (N_{\text{ALQ,Zr}}/W_{\text{ALQ}}) ([^{144}\text{Ce}]_{\text{CS}}/[^{144}\text{Ce}]_{\text{ZR}}) \quad (\text{E-6})$$

Here, we use the subscripts CS and ZR to specify the apparent ^{144}Ce concentrations from the untreated (CS) aliquot, used for determining ^{137}Cs , and the treated (ZR) aliquot, used for determining ^{95}Zr .

1. Derivation of Expressions Describing Bias in the Measured Fission Product Concentrations, $B_R([FP])_{UBT,seg}$

From a Taylor-series expansion of Eq. E-5, the percent relative bias in $[FP]_{ALQ}$ for the ^{137}Cs and ^{144}Ce fission products may be expressed as

$$B_R([FP]_{ALQ}) = B_R(N_{ALQ,FP}) - B_R(W_{ALQ}) \text{ for } FP = ^{137}\text{Cs}, ^{144}\text{Ce} \quad (\text{E-7})$$

Potential bias errors in $N_{ALQ,FP}$, corresponding to the term $B_R(N_{ALQ,FP})$, arose from (1) potential systematic errors in the peak integrations and data manipulations performed by the GeLiGAM software and from (2) potential errors associated with calibration of the detector on the gamma-counting system. In our procedures, the magnitude of $B_R(N_{ALQ,FP})$ was estimated for each fission product from counting data obtained with working standards that were counted along with each set of aliquots. This approach assumed that bias in measuring the number of fission product atoms in each aliquot would equal the bias in measuring the number of atoms of the corresponding fission product in the standards. This assumption may be written as

$$B_R(N_{ALQ,FP}) = B_R(^mN_{STD,FP}) \quad (\text{E-8})$$

where $^mN_{STD,FP}$ stands for the measured value for the number of atoms of the specified fission product in the standard.

Each aliquot weight, W_{ALQ} , was determined with a balance that was calibrated daily using an NBS-traceable standard weight. Bias error in the aliquot weights may, therefore, be considered negligibly small. Consequently, we assign the value zero to $B_R(W_{ALQ})$. Substituting this value and the identity in Eq. E-8 into Eq. E-7, we obtain the expression

$$B_R([FP]_{ALQ}) = B_R(^mN_{STD,FP}) \text{ for } FP = ^{137}\text{Cs}, ^{144}\text{Ce} \quad (\text{E-9})$$

A similar treatment may be applied to Eq. E-6 to obtain an expression for bias in $[^{95}\text{Zr}]_{ALQ}$. In this case, the additional bias error associated with fission product losses in handling the precipitated aliquots must be accounted for. To do this, we add a term, $B_{R,Loss}$, to the expressions describing $B_R(N_{ALQ,FP})$ for the fission product measurements performed on the treated aliquot. Equations E-8 and E-9 express the biases not associated with these losses. The pertinent expressions are

$$B_R(N_{ALQ,Zr}) = B_R(^mN_{STD,Zr}) + B_{R,Loss}(N_{ALQ,Zr}) \quad (\text{E-10})$$

and

$$B_R([^{144}\text{Ce}]_{ZR}) = B_R(^mN_{STD,Ce}) + B_{R,Loss}(N_{ALQ,Ce}) \quad (\text{E-11})$$

Now, by expanding Eq. E-6 in a Taylor-series and substituting appropriate expressions for the variables of interest, we find

$$\begin{aligned}
B_R([^{95}\text{Zr}])_{\text{ALQ}} &= B_R(N_{\text{ALQ,Zr}}) + B_R[^{144}\text{Ce}]_{\text{CS}} - B_R(W_{\text{ALQ}}) - B_R([^{144}\text{Ce}]_{\text{ZR}}) \\
&= B_R(^m\text{N}_{\text{STD,Zr}}) + B_{\text{R,Loss}}(N_{\text{ALQ,Zr}}) + B_R(^m\text{N}_{\text{STD,Ce}}) \\
&\quad - B_R(^m\text{N}_{\text{STD,Ce}}) - B_{\text{R,Loss}}(N_{\text{ALQ,Ce}})
\end{aligned} \tag{E-12}$$

If the fission products ^{144}Ce and ^{95}Zr are assumed to be uniformly distributed in the precipitate, the same fraction of each fission product will be included in the handling losses:

$$B_{\text{R,Loss}}(N_{\text{ALQ,Zr}}) = B_{\text{R,Loss}}(N_{\text{ALQ,Ce}}) \tag{E-13}$$

Equation E-12 is thus reduced to

$$B_R([^{95}\text{Zr}]_{\text{ALQ}}) = B_R(^m\text{N}_{\text{STD,Zr}}) \tag{E-14}$$

This expression for bias error in the aliquot concentration of ^{95}Zr is identical in form to the corresponding expressions (cf. Eq. E-9) for ^{137}Cs and ^{144}Ce . It is important to note that, within counting statistics, the ratio of ^{95}Zr to ^{144}Ce in duplicate precipitated aliquots was observed to be constant, even when the fraction of precipitate recovered for the separate aliquots differed substantially. These observations support the validity of the assumption that underlies Eq. E-13.

The apparent concentration of each fission product in the UBT solution, $[\text{FP}]_{\text{UBT,seg}}$, was computed as the average of the corresponding aliquot concentrations. Since, for the case at hand, the percent relative bias in the average of a group of replicate determinations equals the percent relative bias in each individual member of the group, we may now write an expression for $B_R([\text{FP}]_{\text{UBT,seg}})$ based on Eqs. E-9 and E-14:

$$B_R([\text{FP}]_{\text{UBT,seg}}) = B_R(^m\text{N}_{\text{STD,FP}}) \tag{E-15}$$

We do not attempt to evaluate $B_R(^m\text{N}_{\text{STD,FP}})$ at this time because, as described in Section E.1 below, it cancels when the sum of terms in Eq. E-3 is taken. Thus, its magnitude is of little specific interest.

2. Evaluation of the Variance in $[\text{FP}]_{\text{UBT,seg}}$

When a given fission product concentration in the UBT solution is computed as the average of individual, measured aliquot concentrations, its variance may be estimated from the variance of the measured values as shown in

$$\sigma^2([\text{FP}]_{\text{UBT,seg}}) = \left(\frac{1}{m_{\text{ALQ}}^2} \right) \sum_{\text{ALQ}} \sigma^2([\text{FP}]_{\text{ALQ}}) = \left(\frac{1}{m_{\text{ALQ}}} \right) \sigma_{\text{APP}}^2([\text{FP}]_{\text{ALQ}}) \tag{E-16}$$

In this expression, m_{ALQ} stands for the number of individual aliquot concentrations that were averaged in computing $[FP]_{UBT,seg}$; $\sigma_{App}^2([FP]_{ALQ})$ represents an apparent variance of the measured values and may be estimated from the results of measurements on aliquots from replicate samples. In making this estimate, we pooled the data obtained for a number of different segments to obtain as many degrees of freedom as practical for the estimated variance. In establishing algorithms for our computer programs that carried out the estimate, we wished to pool data over all segments from a given fuel rod. A pooled estimate of the standard deviation of $[FP]_{ALQ}$ could not, however, be calculated in the conventional way because the variance of $[FP]_{ALQ}$ was anticipated to vary from segment to segment owing to variability of the counting statistics obtained for aliquots from any given segment. To overcome this difficulty, the following approach to estimating the variance in $[FP]_{UBT,seg}$ was taken.

From a Taylor-series expansion of Eq. E-5, the variance in each measured value of $[FP]_{ALQ}$ may be described by the relationship

$$\sigma_R^2([FP]_{ALQ}) = \sigma_R^2(N_{ALQ,FP}) + \sigma_R^2(W_{ALQ}) \quad (E-17)$$

In addition, the variance $\sigma_R^2(N_{ALQ,FP})$ may be considered to include contributions from counting errors as well as errors arising from other sources, such as variations in sample geometry, variations in the volume of solution that contained individual aliquots, etc. These variance contributions may be separated in describing $\sigma_R^2([FP]_{ALQ})$ as

$$\sigma_R^2([FP]_{ALQ}) = \sigma_{R,Ctg}^2([FP]_{ALQ}) + \sigma_{R,Oth}^2([FP]_{ALQ}) \quad (E-18)$$

Here, $\sigma_{R,Ctg}^2([FP]_{ALQ})$ stands for the relative variance associated with counting errors for the specified aliquot, and $\sigma_{R,Oth}^2([FP]_{ALQ})$ represents the variance from all other error sources, including variance of the aliquot weight, W_{ALQ} . In our work, the value of $\sigma_{R,Ctg}^2([FP]_{ALQ})$ was available for each aliquot from the error estimates that the GeLiGAM computer programs made during their analysis of each aliquot's gamma spectrum.

If we assume that $\sigma_{R,Oth}^2([FP]_{ALQ})$ has the same value for all aliquots and note that $[FP]_{UBT,seg}$ and the individual $[FP]_{ALQ}$ values for a given segment will be nearly identical owing to the precise nature of our measurements, then Eq. E-18 may be substituted into Eq. E-16 and rearranged to give the relationship

$$\sigma_{R,App}^2([FP]_{ALQ}) = \left(\frac{1}{m_{ALQ}} \right) \sum_{ALQ}^{m_{ALQ}} \sigma_{R,Ctg}^2([FP]_{ALQ}) + \sigma_{R,Oth}^2([FP]_{ALQ}) \quad (E-19)$$

This expression suggests that, although values of $\sigma_{R,App}^2([FP]_{ALQ})$ estimated for individual segments from a given rod could not be directly pooled, they could be used together with the information we had available on counting errors to develop a pooled estimate of $\sigma_{R,Oth}^2([FP]_{ALQ})$. This estimated variance could then be

combined with the counting errors associated with measurements related to each segment to provide our best estimate of $\sigma_R^2([FP]_{UBT,seg})$.

Our computer program was written in a way that first computed the apparent relative variance, $\sigma_{R,App}^2|_{seg}$, of the $[FP]_{ALQ}$ values obtained for each segment in the specified fuel rod, and then computed the relative variance, $\sigma_{R,Ctg}^2|_{seg}$, due to counting errors in measurements related to the corresponding segments. The latter was determined with data from the GeLiGAM output and the formula

$$\sigma_{R,Ctg}^2|_{seg} = \left(\frac{1}{m_{ALQ}} \right) \sum_{ALQ}^{m_{ALQ}} \sigma_{R,Ctg}^2 ([FP]_{ALQ}) \quad (E-20)$$

Variance due to sources other than counting errors was estimated for each segment in the specified rod by taking the difference between the apparent variance and the counting variance, that is

$$\sigma_{R,Oth}^2|_{seg} = \sigma_{R,App}^2|_{seg} - \sigma_{R,Ctg}^2|_{seg} \quad (E-21)$$

Then, the program computed a weighted average of the $\sigma_{R,Oth}^2|_{seg}$ values, using the inverse of the counting variance associated with each segment as the weighting factor for each $\sigma_{R,Oth}^2|_{seg}$ value. This assignment of weighting factors was made because the quality of the $\sigma_{R,Oth}^2|_{seg}$ estimates depended on the magnitude of the counting errors; the larger the variance due to counting for a given segment, the less reliable the estimate. The formula that was used in computing this weighted average is

$$\sigma_{R,Oth}^2|_{Rod} = \frac{\sum_{seg} (\sigma_{R,Oth}^2|_{seg} / \sigma_{R,Ctg}^2|_{seg})}{\sum_{seg} (1 / \sigma_{R,Ctg}^2|_{seg})} \quad (E-22)$$

If the result of this calculation was negative, indicating that the apparent variance for segments from the given rod was smaller than the counting variance, the result was set equal to zero.

Using the value for $\sigma_{R,Oth}^2|_{Rod}$ obtained from Eq. E-22 and the individual values that were obtained for $\sigma_{R,Ctg}^2|_{seg}$ for each segment, the report-generator program calculated a separate estimate of $\sigma_R^2([FP]_{UBT,seg})$ for each segment with the formula

$$\sigma_R^2([FP]_{UBT,seg}) = \frac{1}{m_{ALQ}} (\sigma_{R,Ctg}^2|_{seg} + \sigma_{R,Oth}^2|_{Rod}) \quad (E-23)$$

Although this treatment was based on Eq. E-5, which, strictly speaking, applies only in the case where $FP = {}^{137}\text{Cs}$ or ${}^{144}\text{Ce}$, it works equally well for ${}^{95}\text{Zr}$ measurements if one defines $\sigma_{R,Ctg}^2([{}^{95}\text{Zr}])$ to include counting

errors associated with the ^{144}Ce measurements involved in the ^{95}Zr determination (cf. Eq. E-6). This was done in our ^{95}Zr reporting program by computing $\sigma_{R,Ctg}^2([^{95}\text{Zr}]_{ALQ})$ for each aliquot from the formula

$$\begin{aligned}\sigma_{R,Ctg}^2([^{95}\text{Zr}]_{ALQ}) &= \sigma_{R,Ctg}^2([^{95}\text{Zr}]_{ZRL}) + \sigma_{R,Ctg}^2([^{144}\text{Ce}]_{ZR}) \\ &+ \sigma_{R,Ctg}^2([^{144}\text{Ce}]_{CS})\end{aligned}\quad (\text{E-24})$$

and then substituting these values into Eq. E-20. The subscripts ZR and CS on the right side of this equation correspond to the designations of the precipitated and untreated aliquots from a particular UBT solution sample, just as in the case of Eq. E-6.

To illustrate the magnitude of the random errors associated with $[\text{FP}]_{\text{UBT},\text{seg}}$, we present in Table E-1 a listing of the values we obtained during the EOL campaign for the standard deviations corresponding to $\sigma_{R,\text{Oth}}^2|_{\text{Rod}}/m_{ALQ}$ and $\sigma_{R,Ctg}^2|_{\text{seg}}/m_{ALQ}$. In the latter case, only the value for the segment having the largest associated counting error is listed.

Table E-1. Standard Deviations Associated with Counting Errors and Other Sources in Determining Concentrations of Fission Products in the Unspiked Blend Tank Solutions

Rod No.	Estimated Relative Standard Deviation of the Average of Two Measurements, %					
	Fission Product ^{137}Cs		Fission Product ^{144}Ce		Fission Product ^{95}Zr	
	Counting ^a	Other	Counting ^a	Other	Counting ^a	Other
B	0.21	0.29	0.57	0.40	3.74	0.00
C	0.18	0.15	0.21	0.21	3.01	0.00
D	0.21	0.24	0.29	0.15	4.80	0.00
E	0.14	0.24	0.21	0.40	2.78	0.00
F	0.14	0.17	0.28	0.31	4.56	0.00
G	0.18	0.16	0.32	0.15	5.70	2.09
H	0.14	0.23	0.29	0.24	14.71	0.00
I	0.14	0.35	0.35	0.30	12.17	0.00
J	0.14	0.27	0.25	0.48	7.59	0.00
K	0.14	0.23	0.34	0.14	13.05	0.00
L	0.14	0.17	0.21	0.42	16.41	0.00
M	0.21	0.44	0.35	0.43	97.40	5.23
N	0.21	0.13	0.39	0.26	62.70	3.73
O	0.21	0.09	0.39	0.31	45.33	0.00
P	0.21	0.12	0.35	0.49	66.74	0.00
Q	0.21	0.05	0.46	0.00	62.32	6.92
R	0.28	0.10	0.85	0.45	52.25	0.00

^aValue listed is for segment having largest associated counting errors.

In general, precision of the determinations of UBT concentration for ^{137}Cs and ^{144}Ce was quite good, with each error source contributing a standard deviation of only a few tenths of one percent. Random errors in the ^{95}Zr concentration determinations were considerably larger and were dominated by counting errors. It was recognized, and acknowledged by BAPL, that measurement of the ^{95}Zr fission product monitor would become more and more difficult as the EOL campaign proceeded, because of ongoing decay of this short-lived (half-life of 64.02 days) nuclide. In fact, requirements on the ^{95}Zr determinations were waived as of October 1984 (see footnote b of Table 2 in the report body) with the understanding that ANL was to provide values for this fission product as long as practical, along with reliable estimates of the uncertainties associated with the reported values. Toward the end of the campaign, aliquots that were prepared for the ^{95}Zr measurements were counted for the maximum time (60,000 s) permitted by the gamma-counting system and the counting errors were accepted, regardless of their magnitude. The worst-case counting errors listed for ^{95}Zr in Table E-1 for rods M through R are not representative of all segments processed late in the campaign. Actually, only nine of the 135 segment loadings of ^{95}Zr that were determined at ANL showed counting errors that exceeded 33%, relative.

D. Errors in Counting-Bias Correction Factor, $F_{\text{FP,Rod}}$

The magnitude of the bias correction factor $F_{\text{FP,Rod}}$ was evaluated for each fission product as the average ratio of theoretical-to-measured values for the fission product content of appropriate working standards. The computer program that calculated $F_{\text{FP,Rod}}$ for each fission product provided a separate correction factor for samples from each rod, using data from all standard measurements that were performed on the same days that samples from the given rod's segments were counted. Averaging the apparent counting biases over an entire rod, as opposed to using corrections specific to individual samples or segments, facilitated the calculations and yielded a more-precise estimate of the magnitude of the apparent bias. This approach implicitly assumes that the bias associated with the gamma counting of each fission product remained constant over the time period (two to six weeks) required to process all segments from a given rod. Justification for this assumption is given by our observation that the individual ratios used in calculating $F_{\text{FP,Rod}}$ for each fission product were constant over each rod, within counting statistics, although significant between-rod differences were sometimes found.

The evaluation of $F_{\text{FP,Rod}}$, as described above, may be summarized as

$$F_{\text{FP,Rod}} = \left(\sum_{\text{STD}=1}^{n_{\text{STD}}} \frac{\text{ThN}_{\text{STD,FP}}}{\text{mN}_{\text{STD,FP}}} \right) / n_{\text{STD}} \quad (\text{E-25})$$

In this expression, n_{STD} is the number of standard measurements that were carried out for fission product FP during processing of the specified rod; ThN_{STD} is the theoretical fission product content of an individual working standard; and mN_{STD} is the corresponding measured value for the same standard.

From a Taylor-series expansion of Eq. E-25, the percent relative bias in $F_{FP,Rod}$ may be expressed as

$$B_R(F_{FP,Rod}) = B_R(^{Th}N_{STD,FP}) - B_R(^{m}N_{STD,FP}) \quad (E-26)$$

Bias in $^{Th}N_{STD,FP}$ arose from potential errors in the certified fission-product concentrations for the standardized solution from which the working standards were prepared and from potential bias errors associated with the working standard's preparation. These preparations involved only dispensing of weighed aliquots of the standardized solutions into appropriate counting vials and bringing each aliquot to a predetermined volume in its vial. Each weighing was carried out on a balance calibrated with NBS-traceable standard weights. Radiochemically pure reagents were used for the dilutions. The aliquot preparations are, therefore, considered to have been free of bias.

A limit to the magnitude of potential errors in the certified concentrations of the solutions used for preparing the working standards may be set on the basis of uncertainties quoted on the certificates that describe the solutions. Information pertinent to setting such limits for the ^{137}Cs , ^{144}Ce , and ^{95}Zr standards is given in Table E-2, wherein the standardized fission product solutions that ANL used for the EOL measurements are identified. Taking one-third of the quoted uncertainty for each standardized solution to correspond to its one-sigma uncertainty, we assign the following values to $B_R(^{Th}N_{STD,FP})$ for the three fission products:

$$B_R(^{Th}N_{STD,^{137}Cs}) = 0.16\% \quad (E-27a)$$

$$B_R(^{Th}N_{STD,^{144}Ce}) = 0.43\% \quad (E-27b)$$

$$B_R(^{Th}N_{STD,^{95}Zr}) = 0.43\% \quad (E-27c)$$

Table E-2. Fission Product Standards Used in Calibrating ANL Gamma-Counting Measurements

Fission Product	Source of Standardized Solution	Solution Identification	Quoted Uncertainty	
			Value, %	Conf. Level, %
^{137}Cs	NBS	SRM 4233-B	0.48 ^a	99 ^a
^{144}Ce	Amersham International	Product No. CHZ.72 Solution No. S3/12/6	1.3	99.7
^{95}Zr	Amersham International	Product No. ZCZ.72 Solution No. S3/24/12	1.3	99.7

^aPertains to atom-concentration attribute of standard solution.

Bias in the measured values for each standard was discussed earlier, in Section B.1.a of this appendix; its magnitude is of little interest because it cancels when the product $[FP]_{UBT,seg} \times F_{FP,Rod}$ is taken, as we show in Section E.1 below.

Variance in the counting-bias correction factor, $F_{FP,Rod}$, was estimated in our report-generator computer program as the sum of variances associated with $^{Th}N_{STD,FP}$ and $^{m}N_{STD,FP}$:

$$\sigma_R^2(F_{FP,Rod}) = \sigma_R^2(^{Th}N_{STD,FP}) + \left(\frac{1}{n_{STD}} \right) \Sigma \sigma_{R,Ctg}^2(^{m}N_{STD,FP}) \quad (E-28)$$

In evaluating the terms in this expression, $\sigma_R(^{Th}N_{STD,FP})$ was simply assigned the one-sigma uncertainty of the standards involved in the measurement, i.e., the same values given for $B_R(^{Th}N_{STD,FP})$ in Eq. E-27. Although this assignment may have overestimated the corresponding standard deviations, it was considered a prudent choice in the absence of reasonable alternative estimates.

The second term in Eq. E-28 was evaluated by our computer program using the counting-error estimates that were derived by the GeLiGAM software when each standard measurement was performed. This calculation presumed that variance contributions from all sources other than counting errors were negligible when standards were counted. As we mentioned earlier, no evidence to contradict this presumption was found in the standards data, which conformed to the counting statistics over the duration of operations with any particular rod. Values obtained during the EOL campaign for $F_{FP,Rod}$ and its associated counting errors are presented in Table E-3.

E. Combination of Terms Contributing to Errors in FP_{seg}

1. Bias in the Fission Product Determinations

Bias errors associated with ANL's determination of each fission product may be evaluated by making appropriate substitutions into Eq. E-2. Based on the approximations and definitions presented in the foregoing discussion, particularly Eqs. E-15 and E-26, the percent relative bias in FP_{seg} is given by

$$\begin{aligned} B_R(FP_{seg}) &= B_R(^{m}N_{STD,FP}) + B_R(^{Th}N_{STD,FP}) \\ &\quad - B_R(^{m}N_{STD,FP}) + B_R(W_{UBT,seg}) \end{aligned} \quad (E-29)$$

Because $B_R(^{m}N_{STD,FP})$ cancels in this expression, Eq. E-29 may be simplified to

$$B_R(FP_{seg}) = B_R(^{Th}N_{STD,FP}) + B_R(W_{UBT,seg}) \quad (E-30)$$

Substituting the value 0.02% for $B_R(W_{UBT,seg})$, as discussed in Section B above, using the limiting values for $B_R(^{Th}N_{STD,FP})$ in Eq. E-27, and choosing signs for

Table E-3. Corrections for Apparent Counting Bias and Associated Counting Error Estimates

Rod No.	Factors Used in Correcting for Bias Apparent in Counting Specified Fission Product			Counting Errors Associated with Determining Correction Factor, ^a % Rel. Std. Deviation		
	¹³⁷ Cs	¹⁴⁴ Ce	⁹⁵ Zr	¹³⁷ Cs	¹⁴⁴ Ce	⁹⁵ Zr
B	1.00558	0.99593	1.00289	0.15	0.35	0.12
C	1.00488	0.99295	1.00384	0.11	0.26	0.30
D	1.00532	0.99020	1.00554	0.13	0.31	0.17
E	1.00573	1.00086	1.00651	0.15	0.39	0.13
F	1.00589	1.00417	1.00736	0.17	0.39	0.18
G	1.00684	0.98858	1.00360	0.13	0.25	0.19
H	1.00551	0.98776	1.00601	0.10	0.19	0.16
I	1.00626	0.98493	1.00717	0.13	0.27	0.35
J	1.00732	0.98573	1.00983	0.12	0.23	0.30
K	1.00417	0.99270	1.01289	0.11	0.25	0.56
L	1.00352	0.99671	1.01169	0.12	0.36	0.64
M	1.00570	0.99868	0.99821	0.12	0.29	0.42
N	1.00369	0.99925	1.00494	0.12	0.26	0.33
O	1.00082	1.01357	0.99570	0.13	0.28	0.53
P	1.00014	1.01704	1.01028	0.13	0.31	0.64
Q	0.99791	0.99611	1.00577	0.15	0.36	0.61
R	1.00048	1.00319	0.99745	0.11	0.30	0.31

^aValues correspond to second term on right side of Eq. E-28; these values represent only one of two errors that contribute to the standard deviation of the correction factor. The other error was assigned a relative standard deviation of 0.16% for ¹³⁷Cs and 0.43% for ¹⁴⁴Ce and ⁹⁵Zr.

the individual terms to maximize the value of $B_R(FP_{seg})$, we obtain the following estimates of the maximum probable bias in FP_{seg} for the individual fission product monitors:

$$B_R(^{137}\text{Cs}_{seg})|_{\max} = 0.16\% + 0.02\% = 0.18\% \quad (\text{E-31a})$$

$$B_R(^{144}\text{Ce}_{seg})|_{\max} = 0.43\% + 0.02\% = 0.45\% \quad (\text{E-31b})$$

$$B_R(^{95}\text{Zr}_{seg})|_{\max} = 0.43\% + 0.02\% = 0.45\% \quad (\text{E-31c})$$

These estimates of the maximum probable bias in FP_{seg} for each fission product include only contributions from potential errors in the measurements performed in the individual determinations. They do not include potential bias errors caused by fuel losses that might have occurred during dissolver operations that were carried out for each segment. Such errors are discussed in Section V.C of the report body. Under conditions of normal dissolver operation, fuel losses amounted

to no more than about 0.03%, relative. However, for eight of the 135 fuel-bearing segments, there were mishaps during dissolver operations that increased the potential for losing segment fuel (see Section V.C.3.b of the report body). Values for the maximum probable bias in each fission product determination for each EOL segment were ultimately estimated as the arithmetic sums of the potential measurement biases given in Eq. E-31 and the potential fuel loss biases listed in Table 20 of the report body. These values are summarized in Table E-4.

Table E-4. Potential Bias in Fission Product Determinations

Segment No.	Maximum Probable Bias in Determination of Specified Fission Product, % Relative ^a		
	¹³⁷ Cs	¹⁴⁴ Ce	⁹⁵ Zr
H-08	0.31	0.58	0.58
J-03	0.33	0.60	0.60
J-08	0.25	0.52	0.52
N-02	0.21	0.48	0.48
N-06	0.27	0.54	0.54
O-03	0.43	0.70	0.70
All Others	0.21	0.48	0.48

^aComputed as arithmetic sum of potential measurement bias in Eq. E-31 and potential fuel loss bias from Table 20 in report body.

In every case, the maximum probable bias in the fission product determinations falls within the limits to allowable bias (0.5% for ¹³⁷Cs, 2.0% for ¹⁴⁴Ce, and 2.5% for ⁹⁵Zr) established by project error requirements. Thus, these requirements were met.

2. Standard Deviation of the Fission Product Determinations

The uncertainties we quoted for the segment loadings of each fission product were computed by combining the individual variance contributions from the foregoing discussion according to Eq. E-3. These computations were made with information from calculations based on Eqs. E-4, E-23, and E-28. A separate standard deviation was estimated for each segment because of the appearance of segment-dependent variables in most of these expressions. Use of the project computer system in carrying out the calculations permitted convenient, error-free access to the requisite segment-specific data and ensured that each uncertainty estimate was both realistic and reliable. The percent relative standard deviations estimated for each segment are shown in Tables E-5, E-6, and E-7 for the fission products ¹³⁷Cs, ¹⁴⁴Ce, and ⁹⁵Zr, respectively. For the eight fuel-bearing segments that were involved in dissolver-operation mishaps, additional variance terms were included in estimating the quoted standard deviations to account for uncertainties in (1) any additional measurements that were performed and (2) potentially unrecovered fuel material (see Section V.C.3.b of report body). The standard

deviations given in Tables E-5, E-6, and E-7 for these segments include contributions from these terms.

Standard deviations of the ^{137}Cs determinations ranged from 0.25 to 0.56%, relative, well within the 1.25% limit allowed by project requirements. The ^{144}Ce determinations also met requirements (allowable standard deviation of 2.0%), with estimated standard deviations ranging from 0.49 to 1.10%.

For ^{95}Zr , the October 1984 waiver of requirements exempted values from all rods but B, C, and D from having to meet the 4.0% limit to allowable standard deviation. As Table E-7 shows, this requirement was met for all segments from these rods except segment D-07, which had a low concentration of ^{95}Zr and was consequently exempt from the requirements anyway (see footnote b of Table 2 in the report body). Thus, pertinent error requirements on the fission product determinations were met in every case.

Table E-5. Estimated Standard Deviations of ^{137}Cs Determinations for Individual Fuel Rod Segments

Rod No.	Estimated Relative Standard Deviation for Indicated Segment, %														
	01	02	03	04	05	06	07	08	09	10	11	12	13	14	15
B	0.43	0.41	0.43	0.43	0.43	0.43	0.43	--	--	--	--	--	--	--	--
C	0.32	0.28	0.28	0.27	0.28	0.27	0.29	--	--	--	--	--	--	--	--
D	0.40	0.34	0.34	0.35	0.34	0.34	0.37	--	--	--	--	--	--	--	--
E	0.37	0.35	0.36	0.35	0.37	0.37	0.35	--	--	--	--	--	--	--	--
F	0.34	0.34	0.34	0.33	0.32	0.34	0.33	0.32	--	--	--	--	--	--	--
G	0.35	0.29	0.29	0.29	0.30	0.34	0.29	--	--	--	--	--	--	--	--
H	0.34	0.34	0.34	0.34	0.28	0.34	0.34	0.29	0.34	0.34	0.33	0.32	0.33	0.33	0.32
I	0.44	0.44	0.44	0.44	0.42	0.42	0.44	0.43	--	--	--	--	--	--	--
J	0.38	0.37	0.32	0.36	0.36	0.36	0.36	0.36	--	--	--	--	--	--	--
K	0.35	0.34	0.33	0.33	0.33	0.32	0.34	0.34	--	--	--	--	--	--	--
L	0.31	0.29	0.29	0.29	0.29	0.29	0.29	0.29	--	--	--	--	--	--	--
M	0.52	0.44	0.51	0.51	0.52	0.56	--	--	--	--	--	--	--	--	--
N	0.29	0.28	0.28	0.29	0.30	0.36	--	--	--	--	--	--	--	--	--
O	0.27	0.27	0.32	0.27	0.29	0.35	--	--	--	--	--	--	--	--	--
P	0.28	0.28	0.28	0.28	0.31	0.36	--	--	--	--	--	--	--	--	--
Q	0.26	0.27	0.27	0.27	0.30	0.34	--	--	--	--	--	--	--	--	--
R	0.37	0.28	0.28	0.28	0.26	0.28	0.28	0.28	0.26	0.28	0.28	0.28	0.25	0.25	0.25

Table E-6. Estimated Standard Deviations of ^{144}Ce Determinations
for Individual Fuel Rod Segments

Rod No.	Estimated Relative Standard Deviation for Indicated Segment, %														
	01	02	03	04	05	06	07	08	09	10	11	12	13	14	15
B	0.73	0.74	0.79	0.81	0.81	0.89	0.79	--	--	--	--	--	--	--	--
C	0.58	0.56	0.59	0.58	0.59	0.59	0.59	--	--	--	--	--	--	--	--
D	0.60	0.58	0.60	0.60	0.60	0.60	0.63	--	--	--	--	--	--	--	--
E	0.73	0.72	0.74	0.74	0.74	0.74	0.74	--	--	--	--	--	--	--	--
F	0.70	0.70	0.72	0.71	0.70	0.71	0.69	0.70	--	--	--	--	--	--	--
G	0.59	0.57	0.57	0.57	0.59	0.62	0.57	--	--	--	--	--	--	--	--
H	0.58	0.58	0.58	0.58	0.53	0.58	0.58	0.49	0.58	0.58	0.58	0.58	0.59	0.61	0.58
I	0.63	0.68	0.69	0.68	0.64	0.66	0.69	0.63	--	--	--	--	--	--	--
J	0.72	0.73	0.65	0.72	0.72	0.72	0.72	0.72	--	--	--	--	--	--	--
K	0.57	0.58	0.57	0.57	0.57	0.57	0.62	0.62	--	--	--	--	--	--	--
L	0.74	0.74	0.74	0.74	0.74	0.74	0.74	0.74	--	--	--	--	--	--	--
M	0.78	0.68	0.74	0.77	0.77	0.78	--	--	--	--	--	--	--	--	--
N	0.69	0.68	0.68	0.70	0.68	0.67	--	--	--	--	--	--	--	--	--
O	0.71	0.73	0.72	0.71	0.69	0.70	--	--	--	--	--	--	--	--	--
P	0.82	0.82	0.81	0.82	0.80	0.82	--	--	--	--	--	--	--	--	--
Q	0.66	0.70	0.74	0.68	0.68	0.68	--	--	--	--	--	--	--	--	--
R	1.10	0.76	0.76	0.75	0.75	0.78	0.78	0.75	0.76	0.76	0.78	0.78	0.78	0.78	0.78

Table E-7. Estimated Standard Deviations of ^{95}Zr Determinations
for Individual Fuel Rod Segments

Rod No.	Estimated Relative Standard Deviation for Indicated Segment, %														
	01	02	03	04	05	06	07	08	09	10	11	12	13	14	15
B	2.05	1.21	2.05	1.65	2.19	3.76	3.55	--	--	--	--	--	--	--	--
C	1.54	1.59	1.83	2.02	1.99	2.75	3.06	--	--	--	--	--	--	--	--
D	1.59	1.27	1.34	1.57	1.98	3.37	4.82	--	--	--	--	--	--	--	--
E	1.30	1.37	1.97	2.77	2.46	2.77	2.81	--	--	--	--	--	--	--	--
F	1.99	1.41	2.89	3.23	3.23	4.11	2.74	4.58	--	--	--	--	--	--	--
G	2.56	2.66	3.24	2.77	4.17	4.14	6.09	--	--	--	--	--	--	--	--
H	4.71	1.43	1.94	2.08	1.56	1.43	1.87	1.76	2.08	2.14	2.46	2.74	5.01	4.00	14.72
I	2.10	3.88	4.29	3.78	4.99	6.11	12.18	7.02	--	--	--	--	--	--	--
J	2.09	3.38	4.39	4.95	2.88	4.91	7.61	7.45	--	--	--	--	--	--	--
K	2.21	2.78	4.04	6.20	5.32	8.14	11.70	13.07	--	--	--	--	--	--	--
L	2.99	3.66	5.09	7.51	11.11	8.67	13.78	16.43	--	--	--	--	--	--	--
M	55.10	97.68	11.07	12.02	7.24	5.75	--	--	--	--	--	--	--	--	--
N	60.73	62.92	18.02	13.41	7.68	5.47	--	--	--	--	--	--	--	--	--
O	45.33	23.10	16.30	20.44	8.45	2.82	--	--	--	--	--	--	--	--	--
P	66.74	23.30	31.30	14.58	7.58	3.77	--	--	--	--	--	--	--	--	--
Q	62.71	28.74	28.59	23.23	10.10	8.77	--	--	--	--	--	--	--	--	--
R	52.25	9.95	6.61	10.97	9.12	8.07	8.52	10.88	11.32	14.83	14.30	15.15	41.92	27.74	25.05

ATTACHMENT 1**LIST OF ANALYTICAL PROCEDURES FOR THE
ANL DESTRUCTIVE ASSAY OF LWBR EOL FUEL RODS**

**Analytical Procedures for the ANL Destructive
Assay of LWBR EOL Fuel Rods
August 1984**

	<u>Title</u>	<u>Document No.</u> *
1.	Guide to the Preparation of Reagents for Use in the Dissolution of LWBR Fuel Rod Segments with the Dual Dissolver System (Thorex, Thorex-Aluminum, and Dilute Nitric Acid Rinse)	C-0030-0286
2.	Procedures for the Analysis of Reagents for Use in the Dissolution of LWBR Fuel Rod Segments with the Dual Dissolver System	C-0030-0287
3.	Procedure for Preparation of NBS 993 Spikes for Use in Determining Uranium in LWBR-POB Solution Samples	C-0030-0288
4.	Procedure for Preparation of NBS 950a Spikes (Added to Blend Tank During Dissolution of LWBR-POB Fuel Rod Segments)	C-0030-0289
5.	Procedure for Aliquoting Solution Samples from Dissolved LWBR Fuel Rod Segments under Hot-Cell Conditions	C-0030-0290
6.	Procedure for the Hot-Cell Separation of Uranium from Thorium and Fission Products in LWBR-POB Solution Samples	C-0030-0291
7.	Procedure for Preparing Samples from Dissolved LWBR Fuel Rod Plenum Segments for Measurements of Uranium and Thorium	C-0030-0292
8.	Procedure for Purification of Uranium Prior to Mass Spectrometric Analysis and Preparation of Samples for ^{232}U Determination by Alpha Pulse Height Analysis	C-0030-0293
9.	Procedure for the Determination of $^{232}\text{U}/^{233}\text{U}$ by Alpha Pulse Height Analysis	C-0030-0294
10.	Filament Loading Procedure for Mass Spectrometric Analysis of Uranium	C-0030-0295
11.	Isomass 54R Mass Spectrometer Operating Procedure for Analysis of Uranium in the LWBR-POB Analytical Support Project	C-0030-0296
12.	Procedure for Separation of Cesium Activity from LWBR-POB Dissolver-Solution Aliquots Prepared for ^{95}Zr and ^{144}Ce Determination by Gamma Counting	C-0030-0298

*These documents are available through the Chemical Technology Division, Argonne National Laboratory.

ATTACHMENT 2
CERTIFICATES OF ANALYSES FOR DISSOLVER REAGENTS

LWBR-Proof of Breeding
Analytical Support Project
Argonne National Laboratory

Certificate of Analysis of THOREX Dissolver Reagent

THOREX Reagent Batch No.: 3

Prepared by: R. W. Bane   Notebook Reference: Log 000229, p. 138
Date and Location of Preparation: 11/24/82, D200-B170

Results of Reagent Composition Verification:

Component or Property	Specified Value	Actual Value*	Analyst(s)	ACL Reference
Density	—	1.3711 g/mL	RWB	Log 000229, p. 139
Acidity	13.5 ± 0.2 M	13.51 N	RWB	Log 000229, pp. 138, 139
Fluoride	0.06 ± 0.005 M	0.060 N	RWB	Log 000229, p. 139
Cs Carrier	1.0 ± 0.5 µg/mL	1 µg/mL	EAH	54505 (Spec No.)
Ce Carrier	1.0 ± 0.5 µg/mL	1.4 µg/mL	EAH	54505 (Spec No.)
U Impurities (as ²³⁸ U)	<2 ng/mL	0.3 ng/mL	AME, ELC	NBS Inst. Log No. 8, 12/15/82

*Values determined per procedures in Document C-0030-0287.

LWBR-Proof of Breeding
Analytical Support Project
Argonne National Laboratory

Certificate of Analysis of THOREX-AL Dissolver Reagent

THOREX-AL Reagent Batch No.: 2

Prepared by: R. W. Bane   Notebook Reference: Log 000229, p. 135
Date and Location of Preparation: 11/18/82, D200-B170

Results of Reagent Composition Verification:

Component or Property	Specified Value	Actual Value*	Analyst(s)	ACL Reference
Density	—	1.3763 g/mL	RWB	Log 000229, p. 137
Acidity	13.5 ± 0.2 M	13.44 N	RWB	Log 000229, pp. 136, 137
Fluoride	0.06 ± 0.005 M	0.058 N	RWB	Log 000229, pp. 135, 136
Cs Carrier	1.0 ± 0.5 µg/mL	1 µg/mL	EAH	54489 (Spec No.)
Ce Carrier	1.0 ± 0.5 µg/mL	1.3 µg/mL	EAH	54489 (Spec No.)
Al	0.06 ± 0.005 M	0.06 M	RWB	Log 000229, p. 137
U Impurities (as ²³⁸ U)	<2 ng/mL	0.5 ng/mL	AME, ELC	NBS Inst. Log No. 8, 12/15/82

*Values determined per procedures in Document C-0030-0287.

LWBR-Proof of Breeding
Analytical Support Project
Argonne National Laboratory

Certificate of Analysis of DILUTE NITRIC ACID Dissolver Reagent

DILUTE NITRIC ACID Reagent Batch No.: 1

Prepared by: R. W. Bane Notebook Reference: Log 000229, p. 132

Date and Location of Preparation: 11/3/82, D200-B170

Results of Reagent Composition Verification:

Component or Property	Specified Value	Actual Value*	Analyst(s)	ACL Reference
Acidity	$0.5 \pm 0.05 \text{ N}$	0.477 N	RWB	Log 000229, p. 132
Density	—	1.0116 g/mL	RWB	Log 000229, p. 134
U Impurities (as ^{238}U)	$<2 \text{ ng/mL}$	$<0.1 \text{ ng/mL}$	AME, ELC	NBS Inst. Log No. 8, 12/15/82

*Values determined per procedures in Document C-0030-0287.

ATTACHMENT 3
RESULTS OF LASER CALIBRATION TESTING--
FSS FEED/MEASUREMENT SYSTEM

ARGONNE NATIONAL LABORATORY

30 SOUTH CASS AVENUE, ARGONNE, ILLINOIS 60439

Telephone 312/972-

March 15, 1983

TO: N. M. Levitz Chemical Technology

FROM: D. G. Graczyk *DGG* Chemical Technology
 J. E. Fagan Chemical Technology
 R. Kessie Chemical Technology

SUBJECT: Results of Laser Calibration Testing - FSS Feed/Measurement SystemHighlights

A laser-based displacement monitoring system was used to calibrate and test the performance of the Full-Scale Shear (FSS) feed/measurement system. The test data indicated that the formula used to calculate temperature-corrected position of the carriage on the feed table was incorrect, and that a dislocation in the linearity of the feed system exists, but that reproducibility of the positioning and position-sensing subsystems is exceptionally good.

These observations prompted the modification of the thermal-expansion-correction formula to include coefficients derived from a least-squares analysis of the test data, and the revision of FSS operating procedures to avoid making measurements in regions affected by the dislocation in linearity.

When the new correction formula and operating procedures are applied, the estimated contribution from the FSS feed system measurements to the standard deviation of segment length, boundary location, and fuel rod length determination is only 0.00126 in. Contributions to probable bias is zero for segment length and boundary location determinations, and -0.0003 in. for rod length. To estimate probable errors in the results, these contributions must be combined with those from other error sources. These other errors are, in general, small and confidence is extremely high that end-of-life (EOL) requirements will be met.

Two test rods were measured during the tests. One, a stack of gauge rods previously calibrated by ANL QAD, confirmed the validity of the QAD measurements. The laser system measurement differed by only 0.00034 in. from the temperature corrected QAD value for the 85 in. stack. The second rod was a length of Zircaloy tubing intended for use in the FSS Qualification Test Rod. FSS measurements on both test rods agreed with the laser system values to within 0.0004 in. after adjustment was made for the dislocation bias source, which will be eliminated for future FSS operations by changes in procedure.

I. Introduction

The Full-Scale Shear (FSS) has been developed at ANL as part of the Light Water Breeder Reactor Proof-of-Breeding (LWBR-POB) Analytical Support Project. It is designed to meet DOE-Naval Reactor (NR) requirements for precise location of shear cuts and of clean segment separation for correlation with data from the Production Irradiated Fuel Assay Gauge (PIFAG) under development by Bettis Atomic Power Laboratory (BAPL). The feed system on the FSS comprises a high-precision positioning and measurement system, which will be used for the determination of fuel rod length, segment boundary locations, and segment lengths during the processing of EOL LWBR fuel rods at ANL. This feed system centers around a precision-machined ball screw which drives a carriage upon accurately aligned carriage ways affixed to the FSS machine base. Carriage position on the feed table is indicated by the Absolute Shaft Transducer (AST) display. The AST is a gear-driven counter that accumulates two-hundred counts per revolution of the ball screw. The screw threads are machined such that each revolution provides a carriage travel of 0.2000 in. at a specific reference temperature. Thus, at this reference temperature, each count on the AST indicates a carriage movement of 0.001 in. The digital output on the AST is calibrated to read directly in inches. That is to say, the decimal point in the display is positioned three digits to the left so that an accumulation of 1000 counts will result in a reading of 1.000 on the display. The zero position of the AST readout corresponds to a carriage position at the plane of the cutting edge of the shear blade (the "shear plane").

Carriage movement is accomplished by rotation of the ball screw using a high precision stepping motor (Index Syn), which produces a rotation of 1/200 revolution per stepping pulse. It should be noted that, although both the driving system and carriage position indicator are calibrated to move and detect movement in 0.001-in. increments, the two are completely independent of one another. By design, the Index Syn system is generally the more reliable, allowing the carriage to be positioned with better precision and accuracy than the AST position indicator can measure. Use of the Index Syn readout as a position-indicating transducer is precluded, however, because it provides a measure of displacement only, and loses its point of reference in the event of a power loss. Hence, from the standpoint of recovery-mode operations in the event of a system failure during shearing, the AST, which retains its reading, is the preferred carriage-position transducer.

At temperatures other than the reference temperature for the feed system, thermal expansion of the machine base (including the ball screw) causes a change in the linear equivalent of the AST readout. To accommodate this effect, the machine base temperature is monitored using three resistance temperature sensors (RTDs) attached to the base, and a correction to the inches-per-AST unit calibration is made. This correction is incorporated into a formula used to calculate the actual or true carriage position based on the AST readout.

Mounted on the carriage is another measuring device designated the Rod-End Locator (REL). The REL is a linear variable displacement transformer (LVDT) sensitive to movements as small as 25 μ in. The REL serves several purposes. It is, for example, a useful indicator that the position of a rod end has not moved relative to the carriage during shear operation, as would be

the case in the event of slippage of the carriage clamp jaws holding the rod end. The REL is also an important part of the FSS length measurement system. As indicated previously, the primary indicator of carriage position, the AST, has a limited length resolution due to the 0.001-in. increments to which it responds. By indicating the position of a rod end relative to the plane of the carriage, the REL provides a finely divided scale superimposed upon the "coarse" AST readout. By monitoring both the AST and REL readouts, rod-end positions readable (not necessarily reliable) to 0.00001 in. can be routinely obtained.

Both the AST system and the REL transducer are at their best in measuring displacements rather than absolute positions. For this reason, all length measurements performed with the FSS are made by taking the difference between two carriage positions, each of which is obtained as the sum of the temperature corrected AST reading and the REL value. As a matter of practice, these positions are established as closely as possible using the AST, and the REL reading is restricted to a small range, e.g., ± 0.0005 in. from a suitable reference value. In this way, the temperature-corrected AST value provides a close approximation to the position of interest and the REL supplies only a small correction to the AST-derived value.

The ability of the FSS feed/measurement system to provide reliable rod positioning for segment boundary cuts during LWBR fuel rod shearing and reliable length measurements for whole fuel rods and sheared segments is critically dependent upon the reliability with which these differences in carriage position can be determined. The precision and accuracy of these difference determinations depend, in turn, upon the local and long-range linearity of the ball screw, the validity of the applied temperature corrections, and the reliability of the positioning and readout devices. A series of tests to evaluate the in-cell performance of the FSS feed/measurement system with specific attention to these areas has been completed using a laser-based measurement system. It is the purpose of this report to describe these tests and to present the results and conclusions derived therefrom.

II. Experimental

Performance of the FSS feed/measurement system was evaluated by comparing results from the FSS positioning and readout devices with those obtained with an advanced, laser-based displacement measuring system leased from ELM Systems, Wauconda, IL. The laser system consists of a laser source, an interferometer fixed at an appropriate position on the FSS machine bed, a retroreflector mounted onto the FSS carriage, and a digital readout device. The system provides a measure of the distance between the forward face of the interferometer and the position of the retroreflector. The readout device contains microprocessor circuitry which allows a reference position (zero) to be set at an arbitrary position of the retroreflector relative to the interferometer reference face. It also has the capability of applying automatic corrections for thermal expansion of the machine bed upon which the device is mounted. For our tests, however, it was desired to test the thermal expansion

correction formulas applied to the FSS readout values, and so this feature was not implemented. For these tests, then, the laser system readout corresponded to an absolute distance between the reference position and the position of the retroreflector at the time of reading. The laser-system readout device was interfaced to the Project computer system; a special computer program was written to incrementally advance the FSS carriage, read the output from the laser system and appropriate FSS devices, and store the data for later processing. This computerized data acquisition and subsequent computerized data reduction allowed measurements to be made at a large number of points along the feed system carriage travel.

Two types of measurements were made during the tests: measurements as a function of position on the machine bed, and measurements of the length of specific test rods. These are described separately below.

A. Measurements Along The Shear Bed

For all measurements, the laser interferometer was positioned on the shear bed such that its reference face was as close to the 108.000-in. position of the carriage as could be achieved without interfering with carriage motion. This positioning minimized the effects of relative drift between laser and FSS reference points due to thermal expansion of the FSS machine base. The reference (zero) position of the laser-system retroreflector was then set with the carriage indexed to the position where the FSS Absolute Shaft Transducer (AST) read 108.000 moving the carriage toward decreasing AST values (the forward direction for carriage travel).

For measurements along the length of the shear bed, the carriage was moved to this reference position, the laser readout was reset to zero, and the computer read and stored the following data:

1. The carriage position indicated by the positioning device (Index Syn) on the FSS.
2. The carriage position indicated by the AST.
3. The machine base temperature readouts (needed for thermal expansion calculations), and
4. The laser-system signal (BCD) output.

The computer then indexed the carriage 0.250 in. and read and stored another line of data. This value for the increment of carriage advance was chosen as a compromise between the desire to accumulate a large number of data points and the time available for testing. The computer indexing and reading operations were repeated until AST = 18.000, equivalent to a total carriage travel of 90 in.

The carriage was then jockeyed back and forth several times, to randomize initial conditions, and brought back to the reference position, again approaching the 108.000 point while moving in the forward direction. The laser system zero was reset, and the measurement cycle repeated twice more.

To evaluate possible errors associated with a change in direction of carriage travel, similar measurements were carried out while moving the carriage from AST = 18.000 to AST = 108.000. The reference position for these measurements was maintained at AST = 108.000 for simplicity in comparing results from the two directions of travel. Three separate runs were made in the reverse direction as in the forward direction tests. [NOTE: During the shearing of fuel rods, carriage movement is restricted to the forward direction only. However, in positioning whole, uncut rods for length measurement and initial pre-shearing operations, the rods are moved in the reverse direction. Thus, errors due to backlash during a change in direction of travel are of interest in assessing FSS performance.]

For the purpose of evaluating and "fine-tuning" the formula used for thermal expansion corrections on the FSS measurements, it was deemed desirable to repeat the experiments at a different machine base temperature. The change in temperature, approximately 2°C, was achieved by turning off the cell lights and allowing the temperature to stabilize in this condition overnight. Measurements were made at the lower cell temperature on the following day with the lights still off. Three test runs in each direction of travel were also carried out at this lower temperature.

B. Measurements on Test Rods

The laser system was also used to provide length measurements on several test rods. These measurements were made using the techniques applied in normal FSS Operating Procedures where the carriage position on the shear bed is evaluated with the carriage positioned at the end of a gauge rod of accurately known length, and then at the end of the rod to be measured with both rods positioned against the same fixed stop. The rod length is computed as the sum of the gauge rod length and the difference in carriage positions at the two rod ends.

For the laser-based measurements, the carriage positions at the rod ends were determined with the laser system. Because the carriage can only be indexed in 0.001 in. increments during positioning to the rod ends, small corrections to the laser position values had to be applied using data from the Rod-End Locator (REL) on the carriage. This device is a Linear Variable Displacement Transformer (LVDT) whose readout provides a measure of the location of the rod end relative to the carriage (hence, relative to the laser system retroreflector attached to the carriage). The difference in REL readings (displayed to 0.00001 in.) at the ends of the gauge and test rods is used to interpolate between the 0.001-in. steps taken while positioning the carriage to the rod end.

Two different test rods were measured during the laser calibration tests. The first consisted of a stack of gauge rods (18 to 30 in.) calibrated by ANL's Quality Assurance Division (QAD) and totalling 85 in. in length. The second was a length of power-flattening blanket (PFB) -size Zircaloy tubing intended for use in the FSS Qualification Test Rod.

III. Results and Discussion

A. Preliminary Data Analysis

After data collection was completed, a preliminary analysis of the data was carried out as follows.

First, a statistical analysis of the data from each set of three runs performed under identical conditions (e.g., forward travel with cell lights on) was carried out on a point-by-point basis. This analysis was intended to verify that the reproducibility of the positioning system on the shear feed and of the laser measurements were sufficiently good that measurements made under a given set of conditions were truly representative of those conditions and did not vary significantly when the experiment was repeated. This analysis showed negligible variations among the separate runs at each point. The pooled standard deviation of the difference between the laser reading and the temperature-corrected Index Syn value over the four sets of three runs was only 8×10^{-5} in. As expected, reproducibility of the AST readout at each position between runs was not quite as good. The corresponding standard deviation based on AST values was 5×10^{-4} in., a value consistent with the precision specified by the manufacturer of the AST device.

Having established confidence in the repeatability of the measurements, plots were made of the difference between the temperature-corrected mean AST reading at each point and the corresponding laser reading vs. the mean AST value for each set of conditions. These plots were examined with a view toward identifying trends in the data (see example in Fig. 1) which might indicate ball screw linearity problems, correlation of error with position, or other, similar problems that might limit the reliability of measurements made with the FSS system. This examination pointed out several interesting features:

1. No evidence was found for a difference between data obtained while moving the carriage in the forward direction and that obtained in the reverse direction.
2. A definite trend toward increasing errors with increasing distance from the reference position was apparent (see Fig. 1). This trend was interpreted as an indication that the formula used to correct the FSS measurements for thermal expansion of the machine base was inadequate to its purpose. The results obtained at lower temperature (i.e., with cell lights off) supported this interpretation in that the slope of the error vs. position curve differed significantly between the two sets of conditions.
3. A serious dislocation in the linearity of the feed system was observed at an AST value of about 20 in. (also shown in Fig. 1). This dislocation causes an abrupt shift in carriage position of about 0.0015 in. This shift is important to the performance of the FSS measurement system because it falls between the location of the end of the 30-in. gauge rod originally proposed for use in the measurements and the fuel rod-end. Thus, it would introduce a bias of -0.0015 in. into the fuel rod length measurement.

On the basis of these observations, several revisions to the FSS operating procedures have been implemented. A detailed analysis of the test data, as described in the following discussion, was then carried out assuming the conditions prescribed by these revised procedures.

The procedure changes were twofold:

1. A 35-in., rather than 30-in., gauge rod will be used in measuring the fuel rod length and in gauging the shear plane of the FSS. This longer rod extends beyond the dislocation in the feed system and eliminates its effect on length measurements made with the shear. In the same vein, a longer push rod for use in shearing segments near the fuel rod reference end was designed, so as to preclude the use of carriage positions corresponding to AST value less than 22.0 in.
2. The formula for thermal expansion correction of the AST values obtained from the FSS will be modified to incorporate values for the coefficients as determined by least-squares analysis of the laser test data.

B. Formula for Thermal Expansion Corrections on FSS Measurements

The position on the FSS feed system of the carriage is displayed by the AST readout which measures increments of ball-screw revolution. At a specific reference temperature where the ball screw lead is exactly 0.2000... in./revolution, the AST readout corresponds to the true carriage position in inches. However, at any other temperature, the ball screw lead is different by a factor dependent upon its thermal expansion coefficient and the temperature difference from the reference value. In this case, the revolutions indicated by the AST would correspond to a different position of the carriage on the feed bed. For the FSS feed system, the situation is even more complicated due to construction of the system which compensates, to some degree, differential expansion between the ball screw and the machine base. Thus, an average or effective linear coefficient of thermal expansion for the system as a whole should be used for the calculation. Such a value is difficult to predict *a priori*, and is best determined empirically.

The form of the equation for thermal expansion correction of the AST readout, as used in FSS operations, is

$$P_{\text{CARR}} = \text{AST} (1 + \alpha_{\text{MB}} \cdot \Delta T_{\text{MB}}) \quad (1)$$

where P_{CARR} is the true carriage position relative to the shear plane, AST is the value displayed by the AST, α_{MB} is the linear coefficient of thermal expansion for the feed system, and $\Delta T_{\text{MB}} = (T_{\text{MB}} - T_0)$ where T_{MB} is the average machine base temperature (measured during FSS operation) and T_0 is the temperature at which the AST readings are a true indication of position.

In the laser calibration tests, the laser system readings provide a virtually error-free measure of the distance between the carriage and the reference position. The relationship between the carriage position and AST reading for these tests can be described by the model

$$P_{CARR} = AST (1 + \alpha_{MB} \cdot \Delta T_{MB}) + \epsilon_p \quad (2)$$

where ϵ_p is a randomly distributed variable describing the small deviations of the carriage position about its expected value due to local ball screw nonlinearities, etc. The difference between any two carriage positions is then given by

$$\Delta P_{CARR} = \Delta AST (1 + \alpha_{MB} \cdot \Delta T_{MB}) + \Delta \epsilon_p \quad (3)$$

Rearranging, we may write

$$(\Delta P_{CARR} - \Delta AST) = \Delta AST \cdot \alpha_{MB} \cdot \Delta T_{MB} + \Delta \epsilon_p$$

For the laser calibration tests, the machine base temperature was constant, within very narrow limits, over each set of three repetitive runs; the laser system readings (LSRDG) provide an error-free measure of ΔP_{CARR} , and ΔAST was measured as $(AST_{108} - AST_i)$ for a large number of positions. We may then write

$$(LSRDG - \Delta AST) = \Delta AST \cdot \alpha_{MB} \cdot \Delta T_{MB} + \Delta \epsilon_{108-i} \quad (4)$$

This equation has the form $y = c_1 x + C_2$. A linear least squares analysis was performed on each set of three replicate runs to evaluate C_1 and C_2 . In keeping with the procedural changes described in the previous section, only data points with $AST \geq 22.000$ in. were included in this analysis.

The four values obtained in this way for C_1 are related by the expression

$$C_1 = \alpha_{MB} \cdot \Delta T_{MB} = \alpha_{MB} \cdot T_{MB} - \alpha_{MB} \cdot T_0 \quad (5)$$

This linear relationship between C_1 and T_{MB} for the four experimental C_1 values was also subjected to a linear least squares analysis to obtain values for α_{MB} and the product $\alpha_{MB} \cdot T_O$, from which T_O can be evaluated. Results of the various calculations are shown in Table 1. The appropriate thermal expansion correction formula, based on these results, is

$$P_{CARR} = AST [1 + 1.24 \times 10^{-5} (T_{MB} - 19.56)] \quad (6)$$

This formula will be used in all future operations with the FSS.

Table 1.
Results of Least Squares Analyses of Laser Calibration
Data to Define The Thermal Expansion Correction Formula

<u>Test Run</u> <u>Conditions</u>	<u>Average</u> <u>T_{MB}</u>	<u>$C_1 (= \alpha_{MB} \Delta T_{MB})$</u> <u>$\times 10^5$</u>	<u>$C_2 (\Delta \epsilon_{108}^{-1})$</u> <u>$\times 10^5$</u>
FWD, Lights On	25.79	7.67738	-64.48
REV, Lights On	25.44	7.33416	+56.72
FWD, Lights Off	23.68	5.10115	+37.49
REV, Lights Off	23.80	5.26039	+67.66

$$\alpha_{MB} = 1.24 \times 10^{-5} \text{ in./in.}^\circ\text{C}$$

$$-\alpha_{MB} \cdot T_O = -24.257 \times 10^{-5}, T_O = 19.56^\circ\text{C}$$

C. Errors in Measurements with The FSS Feed System

The errors associated with carriage displacement as indicated by the difference between temperature corrected AST readings for each measurement along the carriage travel are plotted in Figs. 2 through 5 for the various test conditions. The errors were calculated from the relationship

$$AST \text{ ERROR} = (108.000 - AST)[1 + (1.24 \times 10^{-5})(T_{MB} - 19.56)] - \text{LSRDG} \quad (7)$$

Points corresponding to $AST \leq 22.000$ are not included on the plots.

If it is assumed that the errors found at points on the feed system where measurements were made are representative of the errors likely to be observed if measurements were made at all points, then the errors plotted in these figures may be used to estimate the reliability to be expected for length measurements with the FSS. To obtain such an estimate, the error at any position on the feed system is described by the model

$$\text{ERROR} = \bar{\epsilon}_{108-i} + \epsilon_i \quad (8)$$

Where $\bar{\epsilon}_{108-i}$ corresponds to the mean of the differences between the error at $\text{AST} = 108.000$ and that at any other point on the feed system, and ϵ_i is a randomly distributed variable having zero mean and a standard deviation of σ . By virtue of the nature of the least squares analysis performed on the data in order to evaluate the constants in the temperature correction formula, the conditions of this model are fulfilled if $\bar{\epsilon}_{108-i}$ is set equal to the mean of the four C_2 values listed in Table 1. This mean value is 0.000244 in.

The distribution of the random errors about this mean can be described by the variance σ^2 , which is computed as

$$\sigma^2 = \frac{\sum (\text{ERROR} - 0.000244)^2}{(N-1)} \quad (9)$$

where N is the number of data points included in the treatment. This calculation was performed using all 4140 data points having $\text{AST} \geq 22.000$ in. from all four sets of test conditions. The resulting estimate of σ^2 is $(0.000894 \text{ in.})^2$.

During FSS operations in which a segment boundary position is located or a segment length is measured, the difference between two carriage positions is used to provide the measurement. If it is assumed that this difference, when considered over all boundary locations and segment lengths, may fall between any two points on the feed system, then the systematic component of the error, i.e., $\bar{\epsilon}_{108-i}$, will cancel, and the random error will have a variance equal to $2\sigma^2$. Thus, for segment length measurements and boundary locations with the FSS, the feed system will contribute no bias and a variance of $2(0.000894 \text{ in.})^2$ or $(0.00126 \text{ in.})^2$.

In the case of the measurement of a fuel rod length, the assumption regarding the position of the points from which the difference is computed does not hold. In this case, the difference will always be determined from the position of the gauge-rod end, which falls in a relatively narrow range, and the position of the fuel-rod end which falls in the range $\text{AST} = 90.000$ to $\text{AST} = 108.000$. It might reasonably be assumed that the variance associated with positions in these areas of the feed system will be the same as for any other position. However, the mean errors could be significantly different due to local variations in ball screw linearity, etc. To estimate the feed system contributions to the bias and standard deviation of the rod length measurements, the mean error and standard deviation of the

error values were computed for the laser measurements in the range $AST = 22.000$ to 24.000 and in the range $AST = 90.000$ to 108.000 . In the vicinity of the gauge rod end (assuming a 35-in. gauge rod length), the mean error relative to $AST = 108.000$ was found to be -0.00055 in. with a standard deviation, S , of 0.00082 in. For the area of the fuel rod end, the mean error was -0.00027 in. with $S = 0.00079$ in. These data indicate a probable bias contribution to the fuel-rod length from the feed system of -0.00028 in. Since the standard deviations estimated for these regions are smaller than those from the full data set, the contribution to random errors in the fuel-rod length will be estimated as the larger value, 0.00126 in.

In order to project error limits for the boundary location, segment length, and rod length results from the FSS operations, these feed-system error contributions must be combined with contributions from other error sources in the measurements. This combination of errors is beyond the scope of the present report, and will be performed in the context of the Project error analysis. Given the performance of the shear feed system, however, the EOL error requirements should be easily met since the strictest of these stipulate 0.001 -in. allowable bias and 0.005 -in. standard deviation.

D. Measurements on Test Rods

Measurements were made, with the laser system, of the lengths of two test rods. The first of these was a stack of four gauge rods, ranging in length from 18 to 30 in. and having a total stack length of 85 in. From measurements made on the individual gauge rods by ANL's QAD and thermal expansion corrections made using rod temperatures determined during the laser measurements, the stack length was computed to be 85.00858 in. Using the laser system, the stack length at the same temperature was measured as 85.00824 in. The difference between these values, 0.00034 in. is well within the uncertainty associated with the temperature corrected QAD reference value for the stack. This successful cross check between the QAD measurements and the laser system enhances confidence in the accuracy of both measurements. Of the two, the laser-based value is the more reliable. Simultaneously with the laser measurements, the stack length was also determined using the FSS feed system; corrections for thermal expansion of the machine base were applied using the formula derived in section B, above. The stack length was calculated as 85.0063 in., indicating an error in the FSS measurement (relative to the laser measurement) of -0.0019 in. This result is consistent with our earlier discussion since a 30 in. gauge rod was used to gauge the FSS forward stop and, consequently, a -0.0015 in. bias due to the dislocation in the feed system travel is expected in the result.

A similar result was found for a comparison between laser-based and FSS measurements of the length of the Zircaloy tubing intended for the FSS Qualification Test Rod. In this case, the laser measurement gave 116.02825 in. at 25.27°C , while the corresponding FSS value was 116.0265 in. The difference, -0.0018 in., can also be attributed in large part to the effect of the dislocation. If corrections are applied for this effect, the errors are reduced to -0.0004 and -0.0003 in. for the gauge rod stack and Zircaloy tube, respectively. The agreement between the three independent measurements on the gauge rod stack and between the two measurements on the cladding tube are remarkably good.

E. Summary and Conclusions

The laser calibration tests on the FSS feed/measurement system proved both remarkably successful and valuable. The data provided a refined formula for making corrections for thermal expansion of the machine base, allowed estimation of the bias and standard deviation of the feed system measurements, validated gauge rod calibrations by ANL QAD, and provided a length for the Qualification Test Rod Zircaloy-cladding tube. The bias estimates of zero for segment and boundary location measurements and 0.00028 in. for rod length measurements (if a 35 in. gauge rod is used) indicate satisfactorily small error contributions from the FSS feed/measurement system. The standard deviation of an individual position difference measurement is also exceptionally good at 0.00126 in. Even when errors from other pertinent error sources are combined with these values, it is highly improbable that the ECL error requirements on measurements with the FSS feed system will be exceeded.

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3 FORWARD PASSES, AST ERROR

Fig. 1

Error in the Temperature-Corrected Δ AST Value as a Function of Position on the FSS Feed System Before Formula and Procedure Revisions.

FORWARD - LIGHTS ON

Fig. 2

Error in the Temperature-Corrected Δ AST Value as a Function of Position After Formula and Procedure Changes Were Made (Test Conditions are indicated on the Figure).

REVERSE - LIGHTS ON

Fig. 3

Error in the Temperature-Corrected Δ AST Value as a Function of Position After Formula and Procedure Changes Were Made (Test Conditions are indicated on the Figure).

FORWARD - LIGHTS OFF

Fig. 4

Error in the Temperature-Corrected Δ AST Value as a Function of Position After Formula and Procedure Changes Were Made (Test Conditions are indicated on the Figure).

REVERSE - LIGHTS OFF

Fig. 5
 Error in the Temperature-Corrected Δ AST Value as a Function of Position After
 Formula and Procedure Changes Were Made (Test Conditions are indicated on the Figure).

ATTACHMENT 4
REPORTS ON ANL DESTRUCTIVE ASSAY OF LWBR FUEL RODS

Reports on ANL Destructive Assay of LWBR Fuel Rods*

Fuel Rod Serial No.	ANL Letter Identification	Rod Type	Date
2606481	B	PFB	October 16, 1984
2513854	C	PFB	October 25, 1984
2502102	D	PFB	November 13, 1984
2102187	E	PFB	December 7, 1984
2400408	F	PFB	January 7, 1985
2300711	G	PFB	January 25, 1985
3211426	H	Reflector	March 8, 1985
1605519	I	SB	March 14, 1985
1200830	J	SB	May 20, 1985
1302864	K	SB	May 24, 1985
1400544	L	SB	June 12, 1985
0504042	M	Seed	July 12, 1985
0507057	N	Seed	August 13, 1985
0201562	O	Seed	September 11, 1985
0307602	P	Seed	September 12, 1985
0401744	Q	Seed	September 27, 1985
3110505	R	Reflector	November 11, 1985
2514716	W	Performance Verification	June 14, 1985

*These documents are available through the Chemical Technology Division, Argonne National Laboratory.

ATTACHMENT 5
RESULTS OF SINGLE-PELLET PERFORMANCE
TESTING OF THE FULL-SCALE SHEAR

ARGONNE NATIONAL LABORATORY

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August 5, 1982

TO: N. M. Levitz Chemical Technology

FROM: D. G. Graczyk *DGG* Chemical Technology/ACL
 J. Fagan *JF* Chemical Technology
 P. G. Deeken Chemical Technology

SUBJECT: Results of Single-Pellet Performance Testing of the Full-Scale Shear

I. Introduction

The Full-Scale Shear (FSS) has been developed at ANL as part of the Light-Water Breeder Reactor Proof-of-Breeding (LWBR-POB) Analytical Support Project. It is designed to meet Naval Reactors (NR) requirements for precise location of shear cuts and of clean segment separation for correlation with data from the Production Irradiated Fuel Assay Gauge (PIFAG) under development by Bettis Atomic Power Laboratory (BAPL). Continual developmental and characterization testing of the FSS and its components has provided a high degree of confidence that the shear feed system and associated readout devices will be adequate to position and measure LWBR fuel rods to an accuracy and precision in conformance with the stringent NR requirements. Evidence has also accumulated, however, to suggest that the effective plane of separation for fuel material on either side of a cut is shifted from the plane defined by the shear blade edge during each shear stroke. This displacement was designated the "shear-plane error," and efforts were made using a Woods Metal restoration technique to characterize its magnitude and variance in order to evaluate its impact on ANL's ability to meet the requirements relating to clean segment separation during shearing. These early investigations indicated that the magnitude of the shear-plane error might be large enough to jeopardize compliance with existing requirements governing location of the segment boundary-cut position and segment lengths.

The estimates of shear plane error generated during these early tests were based on varying shear test conditions and included contributions attributed to the analytical method used to establish error estimates. Thus, they represented worst case conditions for the magnitude and variance parameters. In spite of the probable overestimation of these parameters, BAPL found that the established Bettis parametric error requirements for destructive segment assay were not exceeded when these estimated shear plane error contributions were added.* [Footnote: *Letter, H. D. Luchinsky to N. M. Levitz, WAPD-LP(CE) POB-425, April 7, 1982.] As a result, it was indicated that the error requirements for ANL measurements of segment boundary location and segment length would be revised and updated to provide sufficient latitude to accommodate shear plane error in the values for fuel material. Established error limits for segment boundary location and segment length for the Zircaloy cladding would, however, be retained.

This declaration by BAPL greatly diminished the urgency of further characterization of the fuel shear-plane error. Other questions related to FSS operations and performance still had to be addressed, however. These included:

- (a) Is there a significant shear plane error associated with the shear stroke through the Zircaloy cladding? Such an effect could conceivably arise from tearing of the cladding edge during hull separation or a rolling over of the exposed cladding end during the shear stroke.
- (b) Is there an advantage to using one or the other of the two blade designs proposed for use with the FSS?
- (c) Is there an advantage associated with shearing at fast or slow speed at segment boundary cuts?

Recognizing that these questions could be answered by the single-pellet shearing tests proposed for a more rigorous evaluation of the fuel shear-plane error, ANL has carried out a series of tests according to that proposal. It is the purpose of this report to describe these tests and to present the results and conclusions derived therefrom.

II. Experimental

The tests consisted of the shearing of simulated fuel rod sections prepared by mounting a single unirradiated ThO₂ ceramic pellet in a short length of Zircaloy tubing. Rods corresponding to three [seed, power-flattening blanket (PFB), and reflector (REF) rods] of the four LWBR rod sizes were used.

A. Preparation of Test Rods

Assembly of the test rods for presentation to the shear is illustrated in Fig. 1. A specially designed brass positioning rod slides into the short Zircaloy cladding sleeve. Two plastic spacers are inserted into the cladding adjacent to the end of the push rod, and the pellet is positioned firmly against the spacers. The cladding length is such that the pellet protrudes from the cladding face approximately 0.02 in..

This test rod configuration was chosen to facilitate operations and data interpretation during the tests. The brass positioning rod provides a means for gripping by the shear feed system clamp jaws. It also serves to align the cladding sleeve, and to support the pellet from behind during rod positioning and shearing. The plastic spacers provide a margin of safety between the pellet and positioning rod so that the positioning rod will not be damaged by the shear blade when the second portion of the pellet is sheared (a simulated boundary cut). The second (boundary) cut is positioned to fall as closely as possible between the two spacers. Protrusion of the pellet from the cladding sleeve allows the reference point of all length measurements on the shear to be the pellet face, and eliminates errors in the fuel shear plane location due to a non-alignment of the cladding edge and pellet face. Similar errors for the cladding shear plane location are not critical as explained in a later section. The same positioning rod was used for all pellets of a given size. Separate pieces of cladding and pairs of spacers were used with a

single pellet for each assembly. Dimensions of the test-rod components are given in Table 1 along with the nominal cut lengths used in shearing the rods as described in a later section.

Table 1.
Dimensions of Test-Rod Components and Nominal
Cut Lengths for the Various Types of Rod Tested

<u>Rod Type</u>	<u>Pellet Diameter,in.</u>	<u>Pellet Length,in.^a</u>	<u>Zircaloy Tube Length,in.^b</u>	<u>Zircaloy Tube O.D.,in.</u>	<u>Segment A Cut Length,in.</u>	<u>Segment B Cut Length,in.^b</u>
REF	0.7415	0.62 to 0.68	3.675 or 3.700 or 3.715	0.832	0.400	0.265 to 0.313
PFB	0.4695	0.36 to 0.38	3.413	0.5265	0.250	0.147 to 0.161
SEED	0.2555	0.45 to 0.47	3.486 or 3.506	0.306	0.150	0.334 to 0.374

^a Length after cutting to remove chamfers, dishes, and corner imperfections.

^b Specific pieces or values chosen to accommodate length of particular pellet used in test rod.

Determination of the lengths of sheared materials was carried out by computing the length equivalent of the material weight. For this procedure to be used, it must be assumed that the linear density of each component is uniform. This assumption is nullified for the thorium pellets by the chamfers, dishes, and corner imperfections on the pellet ends. To overcome this difficulty, the ends of the pellets were cut on a metallographic cut-off saw. The pellets were chucked during sawing to provide opposite faces as nearly parallel as possible.

Before assembling and shearing each test rod, its component parts were weighed and measured. Weights were determined to 0.1 mg. Two aluminum sample cans for collecting the cut portions from each rod were also weighed. Pellet lengths and spacer thicknesses were measured to 0.0001 in. with a digital micrometer. Cladding length was specified with a ± 0.002 in. tolerance

when the cladding was submitted to the ANL shops for cutting. As a check, the individual PFB cladding sections provided by the shops were measured by caliper to 0.001 in. All pieces checked were within tolerance.

B. Shearing Operations

The pellet-bearing section of each test rod was sheared into two segments (referred to as Segment A and B, in Table 1), with each segment collected in a separate aluminum can. The length of the first segment was chosen to correspond to the optimum cut length for the particular rod diameter, i.e., a length-to-diameter ratio of about 0.5. The second was chosen to position the shear cut between the two plastic spacers in the rod. The aluminum cans were capped and the shear blade brushed after shearing each segment. Any weight pickup on the brush was investigated as to the presence of fuel or cladding fines, and material found was included in recovery and material balance calculations.

Two types of shear blade, flat and tapered, were tested. Each blade was used to shear several test rods at both fast and slow speeds (nominally 40 and 2 in./s).

On occasion, while shearing reflector and PFB size rods, a severed cladding hull failed to dislodge from the shear housing into the sample can. With the flat blade, hull hangup was observed only during the course of making the second cut when the cut length was undesirably short and cladding support at the cutting point (provided only by the plastic spacers) was poor. No hulls hung up for cuts of normal length where the support of the cladding is provided by the high density thoria, as in the first portion of the segment. This behavior is consistent with previous observations during shearing where no hulls hung up over hundreds of representative cuts with the flat blade.

In contrast, hull hangup occasionally occurred with the tapered blade for both of the cuts and both shearing speeds. This behavior has been observed with low frequency in previous high-speed shearing tests with the tapered blade, but only rarely in low-speed tests.

No hull hangup was observed for seed-size rods under any conditions.

C. Post-Shearing Measurements

Following the shearing of the test rods, the unsheared cladding section (stub), the plastic spacer in the unsheared rod portion, and the aluminum cans containing the sheared segments were weighed to the nearest 0.1 mg. Each of the aluminum cans was then opened and the contents sorted to separate fuel material, cladding hulls, and the canned spacer. Any cladding fines discovered during sorting were noted, collected, and weighed together with the cladding hulls. The hulls were dusted clean of fuel material and weighed. The plastic spacer collected with the second segment was also weighed separately.

III. Results and Discussion

A. Material Balances

Recovery of the test-rod materials during shearing was monitored by comparing the weights of each type of material recovered with the initial component weights. Recovered cladding was computed as the sum of the stub weight and the hull weights, including metal fines. The weight of fuel in each sample can was computed by taking the full sample can weight minus the cladding hull weight, the empty can weight, and (if present) the plastic spacer weight. The sum of the fuel portions found in each of the two sample cans for a given test rod was compared with the initial weight of the pellet used in the rod.

In general, recoveries of both fuel and cladding were excellent. However, for those reflector rod segments where hull hangup occurred, a disproportionate quantity of fuel was lost. For these cases, the fuel loss was assigned to the segment for which the hull hung up. Data for that segment was then not used in subsequent interpretation and analysis.

Mean values for the fraction of fuel recovered during shearing of each rod type are listed in Table 2, along with the standard deviation of the recovery fractions obtained for each rod. Test rods that showed losses attributed to hull hangup were not included in computing these statistics.

Table 2.
Fraction of Initial Fuel Weight Recovered for
Three Rod Sizes during Single-Pellet Shearing Tests

<u>Rod Size</u>	<u>Mean Value of Fraction of Fuel Recovered, %</u>	<u>Standard Deviation of Recovery Fraction, %</u>
Reflector	99.92	0.14
PFB	99.99	0.06
Seed	99.95	0.34

It should be noted that the data in Table 2 are intended only as an indication of the reliability of the single-pellet test procedures and data treatment and not as an evaluation of the recovery fraction to be expected during EOL shearing operations. Collection of fuel from a single shear cut is expected to be more difficult and therefore poorer than the average collection over a large number of cuts into a given sample can. The best estimate for EOL fuel recovery presently is 99.98% with a %RSD of 0.007% obtained from representative shearing of a large number of unirradiated thoria segments. It is noteworthy, however, that the mean recoveries obtained in these single-pellet tests are sufficient to meet the EOL requirement of 99.75%. The standard deviations of the segment fuel recovery fractions obtained here also compare favorably with the established requirement of 0.25%. The one exception, the 0.34% standard deviation of the fuel recovery fraction for seed-size segments, probably arose from incomplete fuel breakout during shearing of the abnormally long second segment. In some cases, the thoria material had to be manually removed from the cladding hull when the sample can contents were sorted, and overrecovery in these cases sometimes occurred. Since the error contributed by this source was

small in terms of length equivalent of fuel weight, no special treatment was applied to the data obtained.

The objectives of the present tests do not stress weight recoveries of material, but rather deal with length and position capabilities of the shearing system. It is therefore of interest to examine the material balances in terms of fuel and cladding length. Table 3 lists mean values for the length equivalents of fuel and cladding lost during the shearing, collection, and analysis of the test rods. Mean losses of fuel material corresponded to less than 0.001 in. in all cases, with standard deviations (SDs) of 0.0002 to 0.0016 in. Cladding losses were likewise small; the mean losses did not exceed ± 0.0015 in. and the SDs ranged from about 0.001 to 0.005 in. The larger SDs occurred for seed-size rods where the linear densities are low and small weight losses and weighing errors convert to proportionately larger length increments. These results indicate that the single-pellet tests should provide information about the shearing process with a resolution of a few thousandths of an inch.

Table 3.
Length Equivalents of Fuel and Cladding
Lost during Single-Pellet Shearing Tests

<u>Rod Type</u>	<u>Fuel Lost, in.</u>		<u>Cladding Lost, in.</u>	
	<u>Mean</u>	<u>S.D.^a</u>	<u>Mean</u>	<u>S.D.^a</u>
Reflector	0.0006	0.0009	0.0015	0.0017
PFB	0.0000	0.0002	-0.0004	0.0009
Seed	0.0002	0.0016	-0.0012	0.0049

^a Standard Deviation.

B. Evaluation of Fuel Shear Plane Error

The magnitude of the fuel shear-plane error for each test rod was calculated as the difference between the expected segment length (*i.e.*, the length measured on the shear feed system during shearing) and the length actually obtained as determined from the weight of fuel recovered and the pellet linear density. Under the current hypothesis that fuel from the second segment from the pellet (segment B) falls from the sheared fuel face and is collected with the first segment (segment A), segment A will be too long and segment B will be too short by an amount equal to the effective shear-plane error. For the single-pellet shearing tests, the shear-plane error was computed from the data generated on segment A. Values were also computed based on segment B, but these results were used only to verify the segment A calculations.

Mean values and standard deviations of the individual fuel shear-plane error measurements are presented in Table 4 for the various combinations of rod type, blade type, and shearing speed tested. Statistical analysis of these data indicated that, in a few cases, significant differences

exist between the mean shear-plane errors and/or variance of the errors for different conditions. However, from a practical standpoint, the differences are too small to establish a preference for fast vs. slow shearing speed or flat vs. tapered blade design.

Table 4.
Summary of Results for Shear-Plane Errors
Measured for Unirradiated Thoria Fuel Material

<u>Rod Type</u>	<u>Blade/ Speed^a</u>	<u>Mean Shear Plane Error, in.</u>	<u>S.D., in.</u>	<u>No. Tests</u>
REF	F/S	0.018	0.0048	4
REF	F/F	0.021	0.0092	3
REF	T/S	0.024	0.0122	3
REF	T/F	0.023	0.0153	2
PFB	F/S	0.014	0.0004	3
PFB	F/F	0.014	0.0019	3
PFB	T/S	0.018	0.0022	3
PFB	T/F	0.012	0.0014	3
SEED	F/S	0.018	0.0020	3
SEED	F/F	0.014	0.0023	3
SEED	T/S	0.012	0.0031	3
SEED	T/F	0.007	0.0020	4

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^a F = flat; T = tapered; /F = fast; /S = slow.

The data in the table indicate a slightly larger (around 0.020 vs. 0.015 in.) and more disperse (SD of 0.005 to 0.010 in. vs. 0.0004 to 0.003 in.) shear plane error for reflector size rods than for the other rod sizes. It is not clear that this variation is solely due to rod diameter, however, because a different generation of shear tooling was used for the reflector-size rods than for the other two. The earlier design used in manufacturing the reflector-size tooling has been modified

in the new tooling to include changes in dimensions for the blade and housing and in tolerances for the blade/housing gap. The new tooling for reflector size rods was not yet manufactured at the time of these tests. The similarity of results for the PFB- and seed-size rods suggests that the tooling rather than rod diameter has the greater influence on the shear plane error. If this interpretation is valid, then the new tooling design is shown to have merit in these tests.

It is of considerable interest to obtain an estimate of the mean and variance of the shear-plane errors to be used in computing the contributions from this error source to the uncertainties in results obtained during EOL shearing operations. In view of the problems with hull hangup observed for the tapered blade, and since no practical improvement in performance was achieved with this blade design, it shall be established that the flat-bottom blade will be used for EOL shearing. Furthermore, from the standpoint of shear-plane error, fast- and slow-speed operation may be considered identical. In describing the shear-plane error, therefore, data from the flat blade tests at both speeds may be considered to derive from the same population and to represent conditions typical of EOL operations. Thus, the entire set of flat-blade data for each rod type can be justifiably pooled in estimating the fuel shear-plane error. Pooling the data obtained with the different rod types is not as readily justified since the reflector rod behavior is not the same as that for the other rod types. Nevertheless, it can be argued that such an approach will provide a fair and realistic description of the EOL operations as a whole. To compute the "best estimate" of the mean and variance for the fuel shear plane error, then, all of the data obtained with the flat-bottomed blade were pooled, the average taken, and the standard deviation evaluated. The resulting statistics were:

Mean Fuel Shear Plane Error: 0.017 in.

Standard Deviation of Fuel Shear Plane Error: 0.0048 in.

It is interesting to note that a similar treatment of the data obtained in previous experiments to characterize the fuel shear plane error gave a mean of 0.016 in. but a standard deviation of 0.017 in. Thus, the previously determined mean proved to be a good estimate, but the variance included contributions from sources other than dispersion of the shear-plane error. This observation is consistent with statements made with regard to those early tests in which it was believed that the analytical method contributed to the variability of the data.

The values given above for the magnitude and precision of the fuel shear plane errors will be used in the Project Error Analysis to evaluate the shear plane contributions to uncertainties in the segment length and segment boundary location determinations.

C. Evaluation of Errors in Clad Shearing

Deviations between the expected cladding length and the length actually obtained were computed from the weights of sheared clad material in a manner similar to that used for recovered fuel. For each test rod, three different cladding lengths were evaluated. The first was the total length of cladding removed by shearing the Zircaloy tube. It was computed as the linear equivalent of the weight difference between the original Zircaloy tube and the unsheared cladding

stub. The second cladding length was that removed in shearing segment A. It was computed as the linear equivalent of the weight of cladding in the hull collected with segment A. The third cladding length was that removed in shearing segment B as determined from the hull weight for that segment.

To evaluate shearing errors associated with the first two lengths in this list, it was necessary to deduce the amount by which the thoria pellet protruded from the cladding face. This protrusion was computed as the sum of the spacer thicknesses, pellet length, and positioning-rod insertion into the cladding minus the original Zircaloy tube length. All of these parameters had some uncertainty associated with the values used, and error estimates based on these calculations necessarily contained contributions from these sources. It might be anticipated that these uncertainties would combine to give a systematic error in a given set of tests, but would behave in a more or less random fashion between tests. This expectation was taken into account in the analysis and interpretation of the data as discussed below.

For the last length in the list, segment B cladding, both ends of the cladding were sheared after positioning with the FSS feed system. For these cladding segments, the errors estimated from the experimental data should be characteristic of those to be expected from the shearing operations because the errors discussed in the preceding paragraph would cancel.

Errors in each of the three lengths, given by the measured length equivalent of clad weight minus the expected clad length, are summarized in Table 5 for the various test conditions. As in the case of shearing the thoria fuel material, the results from shearing the clad also indicate no practical advantage in using the tapered blade or high-speed shearing to reduce errors introduced during the shear stroke. In fact, the clad data suggest a possible small disadvantage associated with the tapered blade. The larger errors obtained with this blade design might be an indication of tearing or distortion of the cladding edges during the shear stroke and could be related to the problems with hull hangup observed during the tests.

It is noteworthy that the errors listed for the total sheared clad and the clad from segment A are similar in magnitude, direction, and variance for each of the combinations of test variables. As discussed earlier, some portion of these errors arise from uncertainties in the values used to compute the expected length of these cladding segments. However, some error undoubtedly arises from the positioning of the test rods and measurements made on the FSS feed system and from any shear plane error associated with the cladding separation. The errors contributed by these sources can be identified with the uncertainty expected for location of a cladding segment boundary during shearing operations. A limit to the magnitude of these errors can be estimated from the data at hand.

Table 5.
Summary of Results for Errors in
Cladding Length for Various Conditions
in the Single-Pellet Shearing Tests

Rod Type	Blade/ Speed ^a	Mean Error/S.D. Total Sheared Clad, in.	Mean Error/S.D. Segment A, in.	Mean Error/S.D. Segment B, in.
REF	F/S	-0.0018/0.0029	+0.0006/0.0057	+0.0004/0.0036
REF	F/F	-0.0041/0.0015	-0.0051/0.0026	-0.0002/0.0019
REF	T/S	-0.0042/0.0023	-0.0028/0.0053	-0.0032/0.0029
REF	T/F	-0.0106/0.0032	-0.0065/0.0074	-0.0064/0.0056
PFB	F/S	-0.0023/0.0021	-0.0007/0.0035	-0.0012/0.0000
PFB	F/F	-0.0031/0.0020	-0.0012/0.0031	-0.0016/0.0004
PFB	T/S	+0.0022/0.0063	+0.0038/0.0069	-0.0006/0.0003
PFB	T/F	+0.0004/0.0007	+0.0007/0.0011	-0.0003/0.0004
SEED	F/S	+0.0029/0.0065	+0.0047/0.0040	-0.0001/0.0017
SEED	F/F	+0.0056/0.0015	+0.0079/0.0014	-0.0014/0.0005
SEED	T/S	+0.0102/0.0030	+0.0095/0.0029	+0.0051/0.0061
SEED	T/F	+0.0017/0.0010	+0.0098/0.0069	-0.0052/0.0053

^a F = flat; T = tapered; /F = fast; /S = slow.

A limit on the bias in the cladding segment boundary location was estimated as the mean value of the deviation between expected and recovered cladding length for either the total sheared clad or the clad from segment A. This mean was obtained by pooling all of the data from shearing with the flat blade regardless of shearing speed or rod type. Only the flat blade data were used since the tapered blade design has been discounted from consideration for EOL use. The entire set of flat blade data was pooled in anticipation that this treatment would better serve to average out the effects associated with determining the expected cladding lengths. The limiting bias obtained in this way is ± 0.0002 in.

To obtain a limiting estimate on the standard deviation of the cladding segment boundary location, the pooled standard deviation of the data sets used to estimate the bias was evaluated. Again, this treatment was chosen in order to minimize contributions from the experimental uncertainties and provide a realistic estimate of the desired parameter. The value obtained is 0.0035 in. The total estimated error in the cladding segment boundary location, given by the sum of the bias and standard deviation, is thus 0.0037 in. Errors associated with actual EOL operations may be slightly larger than this value due to increased contributions from corrections for thermal expansion of the feed system machine base and cumulative ball screw nonlinearity for long segments compared to the very short segments cut in these tests. Comparison of the values obtained here with the established error requirement of 0.010 in. allowable total error indicates, however, that the requirement should easily be met.

The length of cladding recovered with segment B of each test rod can be used to represent a cladding segment typical of FSS operations wherein each fuel bearing segment will be sheared at both ends. The errors in clad length for the segment B cladding can therefore be used to estimate the bias and standard deviation of EOL cladding segment length with the exception that thermal expansion corrections for the machine base and cumulative nonlinearity of the ball screw will not contribute measurably to the estimates made in this way. Bias in the segment length for Zircaloy clad material was estimated as the mean error in the segment B length, again considering only the flat blade results and pooling data without regard to rod size or shearing speed. The value obtained for this bias is -0.0006 in. Similarly, the standard deviation of the cladding segment length can be estimated as the standard deviation of the segment B clad errors. The value for this statistic is 0.0019 in. These results for the bias and standard deviation of the cladding segment length compare quite favorably with the established error requirements of 0.001 in. and 0.005 in. for the respective parameters.

IV. Conclusions

The single-pellet tests of FSS shearing performance proved successful and valuable. The test results not only provided characterization of the fuel shear-plane error, but also demonstrated the shear's ability to meet established requirements for cladding parameters and resolved the heretofore open questions of blade design and shearing speed to be preferred for EOL operations. It was found that displacement of the effective shear plane during separation of thorium fuel material introduces a bias of 0.017 in. into the cut location and a standard deviation of 0.0048 in. No firm evidence for a significant shear plane error during clad separation was found.

As a result of these tests, use of the flat-bottomed blade at slow speed will be established for EOL operations. No advantage in reducing errors was found with the tapered blade design or with high-speed operation of the shear. Estimated error for shearing segments of Zircaloy cladding material is on the order of 0.004 in. total error (bias plus standard deviation) for the location of the cladding segment boundary, and 0.001 in. and 0.002 in. for the bias and standard deviation, respectively, for the cladding segment length.

DGG:JEF:PGD:lsc

Attach: Fig. 1

Attachment

FIG. 1
TEST ROD FOR SHEAR PLANE TESTS

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