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**VALIDATION WORK TO SUPPORT THE IDAHO NATIONAL ENGINEERING
AND ENVIRONMENTAL LABORATORY CALCULATIONAL BURNUP
METHODOLOGY USING SHIPPINGPORT
LIGHT WATER BREEDER REACTOR (LWBR)
SPENT FUEL ASSAY DATA**

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ABSTRACT

Six uranium isotopes and fourteen fission product isotopes were calculated on a mass basis at end-of-life (EOL) conditions for three fuel rods from different Light Water Breeder Reactor (LWBR) modules and were compared to radionuclide concentrations obtained from chemical assay measurements. The three fuel rods evaluated here were taken from an LWBR seed module, a standard blanket module, and a reflector (Type IV) module. The calculated results were derived using a depletion methodology previously employed to evaluate many of the radionuclide inventories for spent nuclear fuels at the Idaho National Engineering and Environment Laboratory (INEEL). The primary goal of the calculational task was to further support the validation of this particular calculational methodology and its application to diverse reactor types and fuels.

The chemical assay measurements were made by Argonne National Laboratory using destructive examination dissolution techniques. The assay measurements were performed about two years following the LWBR shutdown (1982) on 17 different LWBR fuel rods from four different module types. The rods were sheared into 8-10" segments, dissolved, and analyzed for the six uranium isotopic masses. Three out of the 17 assayed rods were evaluated for the calculation and measurement comparison (one segment for each of the 3 rods). The three fuel rod segments chosen were from (1) a high-zone, high-burnup seed fuel rod, (2) a high-zone, low-burnup standard blanket rod, and (3) a thoria-only, high-burnup reflector rod. The selection of these three fuel rod segments represent a small but diverse sampling of the LWBR fuel rods in the core. Measurement data also included five krypton isotopes, six xenon isotopes, Cs-137, Ce-144, and Zr-95.

The beginning-of-life (BOL), fresh fuel composition in all LWBR Zircaloy-4 clad fuel rods was a binary mixture of uranium and thorium with varying weight percent loadings. The uranium metal was nearly fully enriched in the U-233 isotope (98.2 wt%). Only the BOL thorium reflector modules and regions above and below the active core contained no initial uranium. In addition, to the very complex fuel rod radial loading patterns in the modules, each module also had complex axial loading pattern. The complex loadings patterns served to help flatten the overall core flux power profiles for the axially-asymmetric, moveable, and bottom-inserted seed modules that regulated the core power. The seed movement and time-varying flux profiles added to the difficulty in establishing accurate burnup data.

A calculational validation analysis typically requires the following fundamental input components: (1) a measured energy output (MW-hr, MWd, MWd/MTHM, etc.) or a time-power profile for a specific module, element, rod, or rod segment, (2) BOL rod heavy metal loading, and (3) applicable neutron cross sections. The available LWBR data included everything except a measured energy output (burnup data) for the measured fuel rod segments. Therefore, the fuel rod segment burnups were obtained via an iterative calculational procedure based on matching the measured Cs-137 mass concentration to the calculated Cs-137 mass concentration. In this manner, the LWBR core time-power profile was normalized to the individual estimated segment burnups in order to determine specific time-power profiles for the individual segments needed for the depletion calculation. Comparisons could then be performed between the measured and calculated radionuclides, and the estimated segment burnups with reference rod burnups.

Result comparisons between the calculated and measured mass concentrations in the three rods indicate good agreement for the three major uranium isotopes (U-233, U-234, U-235) with differences <20%. For the seed and standard blanket rod, the U-233 and U-234 differences were within 5% of the measured values (these two isotopes alone represent >97% of the EOL total uranium mass). For the major krypton and xenon fission product isotopes, differences of <20% and <30% were observed, respectively. In general, good agreement was obtained for nearly all the measured isotopes. For those isotopes exhibiting significant differences, possible explanations are

ABSTRACT

discussed in terms of measurement uncertainty, complex transmutations, etc.

The calculated fuel rod segment burnups based on the measured EOL Cs-137 concentrations were estimated to be 37,330 MWd/MTHM, 7,075 MWd/MTHM, and 3,515 MWd/MTHM, for the seed (Rod N segment 3) , standard blanket (Rod I segment 2), and reflector rod (Rod H segment 7), respectively. These burnups are in good agreement with reference burnup data for other module rods in close physical proximity.

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ACRONYMS

ANL	Argonne National Laboratory
ANS	American Nuclear Society
BOL	beginning-of-life
DOE	Department of Energy
ENDF	Evaluated Nuclear Data File
EFPH	effective full power hours
EOL	end-of-life
GWd	Gigawatt-days
INEEL	Idaho National Engineering and Environmental Laboratory
INTEC	Idaho Nuclear Technology Engineering Center
LWBR	Light Water Breeder Reactor
MSID	mass spectrometric isotopic dilution
MW	Megawatt (power)
MWd	Megawatt-days (burnup)
MWd/MTHM	Megawatt-days per metric ton of heavy metal (U + Th)
NBS	National Bureau of Standards
PIE	Post Irradiation Examination
PIFAG	Production Irradiation Fuel Assay Gauge
POB	Proof-of-breeding
PWR	pressurized water reactor
SNF	spent nuclear fuel
TD	theoretical density
wt%	weight percent

1.0 INTRODUCTION

1.1 Purpose

The Idaho National Engineering and Environmental Laboratory (INEEL) has the ownership and responsibility for the storage of many different types of Department of Energy-owned spent nuclear reactor fuels. Understanding the spent fuels through characterization will ensure safe storage and ultimately expedite disposition. Each spent fuel type can be characterized in terms of its basic material properties, geometry, uranium loadings, and accumulated burnup. Further characterization of the spent fuel in terms of its radionuclide inventory can be obtained through depletion or burnup calculations. It is the calculational methodology and the input data used in the burnup calculation that requires some level of validation in order to instill confidence in the accuracy of the calculated radionuclide inventory.

The burnup calculational methodology and input data requirements utilized in the validation work herein are given in some detail in Reference 1. Details of the methodology and requirements are left to the interested reader and will only be partially repeated in this document. The calculational process typically begins by gathering data pertinent to the spent fuel, reactor, and burnup history. Three key blocks of input data that are typically required for a burnup calculation include the following three components: (1) the beginning-of-life fuel element heavy metal loading, (2) fuel element burnup or core power history, and (3) applicable neutron cross sections.

Accuracy or known conservatism in the calculated radionuclide inventory is particularly important from the standpoint that many different engineering disciplines supporting spent fuel activities utilize the inventories as a basis. From the inventories, it is possible to obtain or derive a variety of data that includes: (1) uranium, plutonium, higher order actinide isotopes and concentrations, (2) fission product activities, (3) photon spectra and intensity for gamma source strength (dose rate calculations), (4) activation product activity, (5) activities due to impurities or unusual isotopes not easily measured, and (6) decay heat. In turn, other engineering disciplines will utilize these data for a variety of additional characterization analyses, for example, decay heat for thermal studies and gamma spectra for shielding studies.

One means of estimating the accuracy of the calculated radionuclide inventories is to compare the calculated isotopic masses/activities/decay heats to actual physical measurement data. Physical measurement data typically include quantities such as EOL uranium and plutonium isotopes, decay heat, and occasionally fission product concentrations. Measured data can be obtained from a

variety of destructive and non-destructive techniques which include dissolution chemical assays, mass spectroscopy, and gamma spectroscopy scans. With both calculated and measured data, one can theoretically perform an one-to-one comparison and draw conclusions concerning the calculational accuracy, the fuel burnup, adequacy of the BOL input data, the measurement error, and to a limited extent the measurement technique. Comparison of the calculated EOL nuclide concentrations with measured data is one widely accepted means of validating the calculational method, input data, and ultimately the radionuclide inventory.

As straightforward as the approach seems, simply finding physical measurement data on any particular spent nuclear fuel is at best difficult and often requires extensive research to find useful data. The most frequent situation for INEEL spent fuels is simply that no measurement data exists. Often times, post irradiation examinations (PIE) are performed after the reactor program is finished and is simply not published. However, in the case at hand, the Shippingport Light Water Breeder Reactor (LWBR) program (navy reactor program), a significant amount of useful and well-documented physical measurement data was obtained (references 2, 3, 4, and 5).

Reference 5, in particular, provides a very good overview summary of the LWBR and contains a convenient tabulation of the experimental assay data (original and derived) for the seventeen LWBR fuel rods subjected to destructive chemical assay testing. The derived data are simply a conversion of mole to gram concentrations. The derived data can then be compared directly with the calculated nuclide mass estimates in order to help validate our calculational methodology, establish burnup estimates for the fuel rods, and build confidence in the already calculated LWBR radionuclide inventories.

A direct calculational validation of the LWBR PIE isotopic data was not possible. Burnup data, i.e. energy output for the individual LWBR fuel rod segments was simply not available. In a typical validation analysis, a fuel assembly or fuel element will have a measured total integrated energy output (MW-hr, MWd, MWd/MTHM, etc.) or a time-power profile, beginning-of-life heavy metal loading, and applicable neutron cross sections necessary for a depletion calculation. Data available for the LWBR calculation included everything except the specific rod segment burnup data. Therefore, the fuel rod segment burnup was obtained via an iterative procedure based on matching the measured Cs-137 mass concentration to the calculated mass concentration. In this manner, segment time-power profiles were normalized to these estimated burnups and the other measured isotope concentrations compared to the corresponding calculated concentrations. From these isotopic concentration comparisons, conclusions can be drawn as to the accuracy of the input data, calculational methodology, derived neutron cross sections, and the calculated radionuclide

inventories. The actual calculational procedure employed for the validation effort is discussed in detail below.

The calculations and results documented in this report were performed in support of an ongoing spent fuel validation effort funded by the National Spent Nuclear Fuel Program for the radionuclide inventory and source term development work here at the INEEL.

1.2 The Light Water Breeder Reactor

This section gives a very brief description of the Shippingport reactor, the LWBR program, and the LWBR reactor with specific emphasis on the core and module layout with the intent to inform the reader of the locations of the fuel rods involved in the analyses described herein. A more detailed description the reactor and its operating history can be found in Reference 5 and the extensive references therein.

The Shippingport Light Water Breeder Reactor was a pressurized, light water-moderated and water-cooled thermal breeder reactor. The fuel was binary oxide mixture of high-enriched (98% U-233) urania-thoria oxide fuel. This unique reactor was developed by the Pittsburgh Naval Reactors program to understand and demonstrate the feasibility of **breeding** U-233 with light water thermal reactors, and in addition, improve the nation's utilization of nuclear fuel resources. The LWBR was operated at the Shippingport Atomic Power Station (APS), which was a Department of Energy (DOE) owned reactor plant.

The Shippingport LWBR was successfully operated from August 26, 1977 to December 2, 1982. During the five plus years of operation, the reactor generated approximately 29,047 EFPH (effective full power hours). The reactor power rating was 236.6 MW (thermal). The average bulk coolant temperature was 531 F and operated initially at a pressure of 2000 psia, with reductions to 1940 psia at 4325 EFPH, 1870 psia at 7132 EFPH, and down to 1815 psia at 10,932 EFPH to reduce the probability of fuel rod cladding collapse.

A cross sectional view of the LWBR core is shown in Figure 1. The core consists of four basic modules: (1) seed, (2) standard blanket, (3) power-flattening blanket, and (4) reflector blanket. The three module groups (I-1, I-2, I-3) in the center of the core are symmetrically identical and are each composed of a moveable seed module surrounded by a standard blanket module. These three central seed and standard blanket modules were then surrounded by nine other module groups, each consisting of seed, standard blanket, and power-flattening blanket. The power-flattening blankets were slightly thicker and contained higher U-233 loadings than the blanket regions of the

inner modules. Use of the higher loaded power-flattening blankets produced a relatively uniform power distribution within the interior of the core, thereby simulating the environment of a larger power reactor core. Surrounding the core was a reflector region consisting of thorium reflector modules (two geometry types). The purpose of these reflectors was to both reflect neutrons back into the core and to breed U-233. Figures 2, 3, 4, and 5 show a cross sectional view of the seed, standard blanket, power-flattening, and reflector modules.

The seed, standard blanket, and power-flattening modules all have very complex radial and axial uranium-thorium loadings along with different fuel rod diameters and lattice pitches within each module type. For example, Figure 6 shows the seed and standard blanket radial and axial zone loadings. Although these complex loadings were implemented for good reason, i.e. to optimize the breeding of U-233 and minimize the parasitic neutron absorption in the core, the complex loading arrangements translate directly into greater geometrical complexity in the neutron transport computer model representations needed to capture enough of the important core physics

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Figure 1. Cross sectional view of the LWBR core showing the module types and layout.

	Zone Type	Binary Stack Length	UO ₂ Loading (wt %)	Binary Oxide Theoretical Density (g/cm ³)	Mass Initial Uranium (g/Rod)
High	84"	5.202	10.081	35.87
Low	70"	4.337	10.073	24.89
Low	56"	4.337	10.073	19.91
Low	42"	4.337	10.073	14.93

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Figure 2. The LWBR seed module.

	Zone Type	Binary Stack Length	UO ₂ Loading (wt %)	Binary Oxide Theoretical Density (g/cm ³)	Mass Initial Uranium (g)/Rod
Low	42"	1.654	10.046	19.74
Medium	56"	2.009	10.050	31.97
High	70"	2.739	10.057	54.53
High	84"	2.739	10.057	65.43
Medium	84"	2.009	10.050	47.96

Region 1
261 Standard Blanket Rods (131 top mounted)
0.5712" Diameter
0.630" Pitch

Region 2
303 Power Flattening Rods (151 top mounted)
302 for Types II-1 and Types II-3
0.5265" Diameter
0.630" Pitch

Figure 4. The LWBR power-flattening blanket module.

Thorium Oxide Theoretical Density: 10.03 g/cc
 Percent Theoretical Density: 97.28%
 Stack Length: 104 inches
 Initial Mass Thorium: 6,310.5 g/rod

Identification legend example:

Designates top or bottom mounted
 1 - bottom mounted
 2 - top mounted
 Designates numerical position in row
 Designates row

228 rods (113 top mounted)
 0.832 dia.
 0.900 pitch

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Figure 5. The LWBR reflector module (Type IV).

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Figure 6. Axial cross sectional view of the seed and standard blanket modules showing the complex axial and radial rod loading of uranium and thorium.

phenomena in order to ultimately produce an accurate depletion calculation and radionuclide inventory. It must be remembered that our goal on the spent fuel programs is not necessarily to perform a full fledged reactor core physics analysis for each and every fuel type and element, but rather to perform a cost- and time-effective analysis that provides sufficient accuracy to ensure reasonable and conservative estimates of the radionuclide inventories.

Figure 7 shows the locations of LWBR fuel rods that were inspected and/or chemically assayed. The three fuel rods selected for the validation effort in this study are indicated on Figure 7 and individually on Figure 2 (seed module), Figure 3 (standard blanket module), and Figure 5 (reflector module). Radionuclide inventories were calculated for a single, specific segment from each of these three fuel rods and were then compared with the corresponding measured data.

1.3 LWBR Assay Measurement Data

Chemical assay measurement data, as mentioned above, exists for select LWBR fuel rods. All together seventeen (17) individual LWBR fuel rods from different seed, blanket, and reflector modules were cut up and analyzed. The physical measurement data for the 17 LWBR seed, blanket, and reflector fuel rods are well documented in the destructive assay reports obtained through personal communication with Argonne National Laboratory (ANL). The ANL reports are available in individual reports or one can find the data for all seventeen rods conveniently tabulated and summarized in reference 5 (Appendix D).

Table 1 below lists the alphanumeric identification, LWBR module, module location, and the number of shear segments for the 17 assayed fuel rods. Note: the module location is a letter designation followed by a number; the letters refer to the hexagonal ring in the module and the numbers refer to the position in the ring (see Figures 2-5).

The chemical assay measurement process first started by shearing each of the seventeen 104" long, zirconium-clad fuel rods into approximately 8-10 individual axial segments of approximately equal length. The oxide fuel material in each segment was then dissolved and analyzed. The specific isotopes analyzed for include:

- Six **uranium** isotopes: U-232, U-233, U-234, U-235, U-236, and U-238,
 - Five **krypton** fission product isotopes: Kr-82, Kr-83, Kr-84, Kr-85, and Kr-86
 - Six **xenon** isotopes: Xe-128, Xe-130, Xe-131, Xe-132, Xe-134, Xe-136,
- Three individual **fission product** isotopes: Cs-137, Ce-144, and Zr-95.

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Figure 7. LWBR core cross section showing the locations of all the fuel rods subjected to inspection and/or chemical assay and the three individual fuel rods selected for calculation and comparison to assay data.

Table 1. The seventeen destructively assayed LWBR rods.

Rod ID	Module	Location	Shear Segments
B	Power-flattening blanket	E31	9
C	Power-flattening blanket	F73	9
D	Power-flattening blanket	H1	9
E	Power-flattening blanket	B62	9
F	Power-flattening blanket	C13	10
G	Power-flattening blanket	D29	9
H*	Reflector IV	B1	17
I*	Standard blanket	E56	10
J	Standard blanket	A49	10
K	Standard blanket	D24	10
L	Standard blanket	C3	10
M	Seed	L29	8
N*	Seed	C10	8
O	Seed	P39	8
P	Seed	N63	8
Q	Seed	M49	8
R	Reflector	E3	17

The three asterisked rod identification letters (H, I, and N) are the rods evaluated in this report.

The two xenon isotopes, Xe-128 and Xe-130, are not fission products, but rather the daughter products resulting from the radiative capture and subsequent beta-decay of the fission products I-127 and I-129, respectively. Hence, because Xe-128 and Xe-130 are dependent on the time-varying concentration and capture cross section of a fission product, the concentrations of these two isotopes are much more difficult to accurately calculate than the other krypton and xenon direct-yield or beta-decay chain fission products. Also, the two fission products, Ce-144 and Zr-95, are radioactive and have relatively short half-lives (285 days and 64 days, respectively).

The main purpose of the post irradiation examinations was to evaluate the breeding efficiency of the LWBR. The assay data provided an estimate of EOL U-233 concentrations in selected fuel rods which were then compared with neutronic calculations that provided an estimate of the total full core U-233 core inventory at EOL. In addition to the U-233 concentration, the assay data also provides concentrations for the full range of the uranium isotopes (U-232, U-233, U-234, U-235, U-236, and U-238) which are very useful for validation comparison. Plutonium and higher order actinide concentrations were not measured, probably because of their low production concentrations.

One piece of information vital to the validation effort that was not included in the assay measurement data was a value for the fuel rod burnup (MWd/MTHM). A validated rod segment burnup value corresponding to each segment or rod would have greatly simplified our calculations, because it would have allowed us to fix the burnup, perform the depletion calculation, and simply compare the calculated isotopics with those measured. However, because the rod segment burnups (MWd/MTHM) were not given, the segment burnups had to be based on the measured EOL Cs-137 concentration.

In the calculation, in order to match the Cs-137 concentration to the measured value, a burnup (normalized power level) was initially estimated and the depletion calculation performed and the concentrations compared. The burnup was then continually adjusted or renormalized until the concentrations were in good agreement. This iterative procedure resulted in a calculated estimate of each of the three fuel rod segments burnup in terms of MWd/MTHM which are reported herein.

In a sense, we are really estimating a burnup for the fuel segments by iteratively changing the burnup (MWd) over the LWBR power history until the calculated and measured EOL isotopic concentrations are equally matched or sufficiently converged. The Cs-137 isotope was chosen for several reasons: (1) good EOL measurement or assay data were available, (2) Cs-137 as a fission product has a relatively large fission yield and results in relatively high EOL concentrations in the

fuel rods, and (3) Cs-137 has a relatively long half-life (30.17 years) and basically accumulated over the burnup of the LWBR core and is then one of the most reliable isotopes (of the isotopes measured) for estimating fuel rod segment burnups.

The U-233 isotope can also be used in the burnup estimate process, because of its relatively high EOL concentration and excellent measured accuracy. However, the U-233 concentration is not as straightforward to calculate because of the complex and competing processes governing the destruction and breeding of this isotope relative to a high yield and direct beta-decay chain producing a fission product such as Cs-137. In every fuel rod, the U-233 concentration varies as a function of burnup time due to competing processes of destruction via the fission, radiative capture, (n,2n) reactions, etc.; and production due to transmutation from Th-232 and the (n,2n) reaction from U-234. Fuel rod location in the core also effects the U-233 loading (flux level, spectral effects, module leakage, burnup, etc.) along with the initial fuel rod uranium loading. From a calculational standpoint, one might expect estimates of the EOL U-233 concentration to be less accurate because of the additional dependency on other isotopic concentrations and burnup-dependent reaction cross sections needed in the calculation. In other words, there are simply more variables involved in the calculation that effect the EOL U-233 concentration versus the Cs-137 concentration.

The calculational validation effort focused on a single segment from each of the three different fuel rods for the depletion analysis and radionuclide inventory estimates:

(1) The first fuel rod (**Rod N**) selected was from a seed module near the very center of a module, i.e. a very high burnup region of the module. The segment chosen was Segment 3, which included the axial or midplane of the rod and would have experienced close to the maximum burnup for any seed fuel rod.

(2) The second fuel rod (**Rod I**) selected was from a relatively low burnup standard blanket fuel rod near the outer periphery of the blanket in the high zone region. The segment chosen was segment number 2 located at the very top of the active fuel rod region and just below and adjacent to the top axial thorium reflector. This segment was specifically chosen because the burnup was expected to be relatively low and because it was located on the axial thorium boundary (i.e. very different conditions relative to Rod N, Rod I was selected to test the versatility of the calculational methodology).

(3) The third rod (**Rod H**) selected was from a reflector module (Type IV). This rod was located in the first row adjacent to the LWBR core and would have experienced near maximum burnup for reflector rods and hence maximum EOL concentrations of bred U-233.

The three different rod segments were carefully selected in order to vary particular effects and test the calculational methodology. The intent was to ultimately validate the methodology and its robustness along with the associated radionuclide inventories.

In the case of the LWBR, three important input data fields were required to accurately predict the radionuclide inventory of the individual rod segments, namely, (1) the reactor operating history (power production periods and shutdowns), (2) the Beginning-of-Life (BOL) fuel rod uranium and thorium loadings, and (3) the neutron cross sections as a function of burnup over the core operating lifetime. The following three sections describe in greater detail the three fundamental input data fields.

1.4 LWBR Operating History

The operating history of the LWBR is given in Reference 10. Average daily generator electrical output from the LWBR plant is provided in this reference from which a detailed power history for use as input in the depletion calculation can be developed. For the validation work, the average quarterly thermal core power outputs based on 3-month increments over the total LWBR operating history were used to simplify the amount of input.

As an example, the power profile for an average seed module is given in Table 2. The time periods are given in 3 month or approximately 90-day increments. In addition, the Type I Seed module thermal power output is estimated for that operational period along with the burnup in MWd (megawatt-days). The last two columns of the table are estimated average and maximum power levels for Rod N segment 3 based on two different accumulated burnups used in the burnup iteration process. These power profiles were continually re-normalized (adjustment of the burnup) until the calculated EOL Cs-137 mass concentration was equal to the measured EOL Cs-137 mass concentration.

1.5 BOL Uranium and Thorium Fuel Rod Loadings

The LWBR is an extremely complex core from the standpoint that there are a variety of module types, fuel rod types, fuel rod diameters, fuel rod axial loadings, and module radial loadings. Despite the complexity, the **BOL fuel element loadings** for each fuel rod type are readily available in several reports, most notably reference 11. Data from this reference were used to establish the BOL fuel number densities and BOL uranium and thorium mass loading for all three of the rod segments evaluated.

Table 2. Seed module power history by operating quarter.

Reactor Operation START (date)	Reactor Operation END (date)	Duration (days)	Single Type I Seed (MW(th))	Single Type I Seed (MWd)	based on segment mass MWd/MTM Cumulative (days)	AVERAGE 4.2046 MWd 27750.00 Seed Type 1 Rod N Segment 3 (MW(th))	MAXIMUM 6.2425 MWd 41200.00 Seed Type 1 Rod N Segment 3 (MW(th))
26-Aug-77	30-Sep-77	35	2.731274	95.5946	35.000	1.1183E-03	1.6603E-03
30-Sep-77	31-Dec-77	92	5.971203	549.3507	127.000	2.4449E-03	3.6298E-03
31-Dec-77	31-Mar-78	90	7.897081	710.7373	217.000	3.2334E-03	4.8006E-03
31-Mar-78	30-Jun-78	91	5.969611	543.2346	308.000	2.4442E-03	3.6289E-03
30-Jun-78	30-Sep-78	92	6.767031	622.5668	400.000	2.7707E-03	4.1136E-03
30-Sep-78	31-Dec-78	92	7.217013	663.9652	492.000	2.9549E-03	4.3871E-03
31-Dec-78	31-Mar-79	90	7.549443	679.4499	582.000	3.0910E-03	4.5892E-03
31-Mar-79	30-Jun-79	91	0.000000	0.0000	673.000	0.0000E+00	0.0000E+00
30-Jun-79	30-Sep-79	92	5.202659	478.6447	765.000	2.1302E-03	3.1626E-03
30-Sep-79	31-Dec-79	92	6.666736	613.3397	857.000	2.7296E-03	4.0526E-03
31-Dec-79	31-Mar-80	91	5.068303	461.2156	948.000	2.0752E-03	3.0810E-03
31-Mar-80	30-Jun-80	91	5.999137	545.9215	1039.000	2.4563E-03	3.6468E-03
30-Jun-80	30-Sep-80	92	6.289765	578.6584	1131.000	2.5753E-03	3.8235E-03
30-Sep-80	31-Dec-80	92	2.911629	267.8699	1223.000	1.1921E-03	1.7699E-03
31-Dec-80	31-Mar-81	90	6.354118	571.8706	1313.000	2.6016E-03	3.8626E-03
31-Mar-81	30-Jun-81	91	3.731881	339.6012	1404.000	1.5280E-03	2.2686E-03
30-Jun-81	30-Sep-81	92	6.338568	583.1482	1496.000	2.5953E-03	3.8531E-03
30-Sep-81	31-Dec-81	92	5.039344	463.6196	1588.000	2.0633E-03	3.0634E-03
31-Dec-81	31-Mar-82	90	5.325345	479.2810	1678.000	2.1804E-03	3.2372E-03
31-Mar-82	30-Jun-82	91	5.670859	516.0482	1769.000	2.3219E-03	3.4473E-03
30-Jun-82	30-Sep-82	92	5.467423	503.0029	1861.000	2.2386E-03	3.3236E-03
30-Sep-82	2-Dec-82	63	0.031986	2.0151	1924.000	1.3096E-05	1.9444E-05

SUM= 1924 Total MWD= 10269.1357 MWd
Total U+Th metal mass*= 370063.1200 gm
Burnup= 27749.69 MWd/MTHM
Burnup= 6.48E+20 fiss/cc

* MTHM=U+Th metal mass in UO₂+ThO₂ binary pellets and ThO₂ steps, but not the top and bottom ThO₂ reflectors.

Decay START (date)	Decay END (date)	Duration (days)	(MW(th))	(MWd)	Cumulative (days)
2-Dec-82	2-Jul-83	212	0	10269.1357	2136.000
2-Jul-83	2-Dec-83	153	0	10269.1357	2289.000
2-Dec-83	2-Jul-84	213	0	10269.1357	2502.000
2-Jul-84	2-Dec-84	153	0	10269.1357	2655.000
2-Dec-84	16-Jul-85	226	0	10269.1357	2881.000

SUM= 2881

Included in the reference data are the following: (1) UO_2 weight percent loading in the various module binary oxide zones (low, medium, high), (2) uranium isotopic enrichment in weight percent (i.e. U-233 (98.23 wt%), U-234 (1.29 wt%), U-235 (0.09 wt%), U-236 (0.02 wt%), and U-238 (0.37 wt%)), (3) urania and thoria theoretical densities (TD) and the actual oxide fuel fabrication density, (4) axial zone lengths, and (5) BOL fuel pellet diameters. Figures 2, 3, 4, and 5 include summary tables of this data and additional derived data used to develop BOL uranium and thorium number densities and mass loading for the various module zones and fuel rods. Note the data given in the summary tables in Figures 2, 3, 4, and 5 are the data that were used in the calculations here (minor variations in the data exist relative to the same data given in reference 5).

Reference data also includes elemental impurities in the oxide fuel compositions, however, these oxide fuel impurities are not included in the validation depletion calculations, primarily because the measured data does not include isotopic mass concentrations other than the uranium isotopes and selected fission products.

1.6 Neutron Cross Sections

The third data component required for the depletion calculation is the burnup-dependent **neutron cross sections**. Utilization of appropriate reactor-specific neutron cross sections in a burnup calculation is vital, particularly for actinide transmutation. Not only do cross sections vary from reactor to reactor, but they vary spatially within a given reactor and with burnup. Due to these complexities and the difficulty in developing cross sections, one must be very selective in choosing appropriate neutron cross sections for the depletion calculation.

The ORIGEN2 depletion computer code was used to perform the burnup calculation in the validation methodology. This code comes with several standard neutron cross section libraries, however, these libraries have been developed for specific reactors and fuels at various burnups. Some of these standard libraries are mentioned in section 2.0 (see Reference 1 for a more detailed discussion of the standard ORIGEN2 libraries). In general, these libraries are not applicable for the typically non-standard research and test reactors with unique neutronic designs and spectral properties. For these unique reactors it is highly recommended that new reactor-specific neutron cross section libraries be developed per the methodology described in section 3.0

To expedite the burnup calculations, often times it is possible to use a previously generated cross section library if there is enough similarity between reactors and fuel type. It is always prudent to first assess the effects of a variety of factors that influence the cross section magnitude, for example:

- (1) fissile loadings,
- (2) lattice pitch,
- (3) rod diameter,
- (4) cladding type,
- (5) water-to-metal ratio,
- (6) reactor moderator/coolant
- (7) burnup,
- (8) fuel element poison loading,
- (9) reactor or element reflectors
- (10) strong absorbers (poison rods and control rods)

In general, however, cross section development for unique research and test reactors becomes a necessity for good radionuclide inventory estimates. Since there are no standard LWBR ORIGEN2 neutron cross section libraries, reactor specific cross sections had to be developed for the validation effort here.

2.0 COMPUTER CODES

The following three reactor physics computer codes form the basis of the calculational burnup methodology and are used to calculate the reactor-specific cross sections and perform the fuel rod depletion calculations. The codes are used together in an iterative manner to achieve the desired level of burnup and accuracy. A brief description follows along with references that provide more details.

2.1 MCNP

The **MCNP4B** (Monte Carlo N-Particle) code version 4B (reference 6), is a general purpose, continuous energy, generalized geometry, coupled neutron-photon-electron Monte Carlo transport code. The neutron energy range is from 10^{-5} to 20 MeV, the photon energy range is from 1 keV to 100 MeV, and the electron energy is from 1 keV to 1000 MeV. Tally quantities include particle or energy current and flux across a surface; and, flux, energy deposition, and reaction rates averaged over cell volume(s). A wide variety of nuclide cross sections and reactions are available from the ENDF-4, ENDF-5, and ENDF-6 data sets for tally purposes.

2.2 MOCUP

The **MCNP-ORIGEN2 Coupled Utility Program** code or MOCUP (version 7) is a system of external processors that links input and output files from the MCNP4B and ORIGEN2.1 codes in order to perform a time-dependent burnup of a fuel composition. MOCUP is composed of three processing modules, namely, mcnpPRO, origenPRO, and compPRO. Each module performs a specific, sequential task during each burnup iteration. No modifications are required to either the MCNP or ORIGEN2 codes in order to run in the MOCUP code system. MOCUP operates strictly on the MCNP and ORIGEN2 input and output files.

2.3 ORIGEN2

The **ORIGEN2** code version 1 (reference 8 and 9) is used for calculating the buildup, decay, and processing of radioactive isotopes. Several standard libraries of cross section, decay, and production data are available with the code to allow for tailored applications. ORIGEN2 will output concentrations, activities, and decay heats for over 1700 activation products, actinide and daughter products, and fission product isotopes. A special feature in ORIGEN2 is a user option to modify the standard cross section libraries (base library) with user calculated cross sections, thereby allowing a burnup calculation to be more reactor specific. ORIGEN2 performs its depletion calculations using effective one-group cross sections with depletion controlled either by

total power or by the irradiation flux level. The matrix exponential method is used to solve the large numbers of coupled differential equations describing the complex burnup and decay mechanisms between the nuclides.

2.3.1 ORIGEN2 Standard Libraries

The ORIGEN2 code relies on nuclear data libraries that supply neutron cross sections, photon cross sections, radionuclide photon and neutron emission spectral intensities, radionuclide half-lives, fission product yields, etc., i.e. data needed for a depletion calculation. Data for each stable and radioactive nuclide is contained in these libraries. Of particular interest is the neutron cross section data. This data is problem-dependent and requires special development for each specific problem as in the case of the LWBR. All other library data remains unchanged throughout the depletion calculation. The burnup methodology in section 3 will describe how specific neutron cross sections are generated for the three different LWBR fuel rods.

In the past, data libraries have been developed for various reactor types, particularly for commercial reactors that would typically have a greater user base relative to the one-of-a-kind reactors typical of many of the spent nuclear fuels stored at the INEEL. The "commercial" data libraries that come with the ORIGEN2 code are referred to as the standard libraries. In general, one will start with a standard library as a "base" library and modify as many of the nuclide neutron cross sections as appropriate for the particular problem and application.

Standard cross section libraries exist for several different initial enrichment and burnups in typical PWR fuel cycles. The standard library (pwrxslib) selected for the "base" case was derived from a PWR core with an initial enrichment of 3.2 w/o and standard discharge burnups. This "base" library was updated as a function of burnup over the LWBR power cycle.

All raw cross section data used in the cross section generation methodology was derived from ENDF/B-IV, ENDF/B-V, and ENDF/B-VI neutron cross section data and processed with the codes from the AMPX package and then again processed by the NJOY code for usage by the MCNP code.

The standard photon library for ORIGEN2 contains data for five different sources of photons: (1) x-rays and gamma rays from radioactive decay, (2) gamma rays from (α ,n) reactions, (3) prompt fission gamma rays from spontaneous fission, (4) gamma rays from fission products produced during spontaneous fission; and (5) bremsstrahlung. The radioactive decay and bremsstrahlung data were generated from information in the Evaluated Nuclear Structure Data File and the

remaining three sources calculated from empirical relations. Note: The standard photon library (pholibuo2b) used in the LWBR calculations was not modified in any way.

The standard decay library for ORIGEN2 contains half-life, decay mode, recoverable energy per decay, natural abundance, and allowable concentrations in air and water for each nuclide. This data is presently available for over 1700 different isotopes. By its nature, this information is completely independent of the nuclear system being considered, and thus can be used for all reactor types. These values were painstakingly gathered from the Evaluated Nuclear Structure Data File with supplements from ENDF/B-IV and ENDF/B-V. Calculational comparisons with the ANS-5.1 decay heat standard were in excellent agreement for most decay times longer than about 50 seconds. Note: The standard decay library used in the LWBR calculations was not modified in any way.

3.0 BURNUP METHODOLOGY

The burnup calculation uses three computer codes, namely, MCNP, MOCUP, and ORIGEN2. The MCNP code performs the neutronic transport calculation to determine the cell flux and reaction rates that translate into neutron cross sections; the ORIGEN2 code performs the time-dependent, nuclide coupled depletion or burnup calculation to obtain radionuclide concentrations; the MOCUP code is simply a utility code that links MCNP and ORIGEN inputs and outputs.

The ORIGEN2 code with its accompanying standard libraries can be used to perform a burnup calculation. However, if these standard libraries are not applicable to the reactor and reactor fuel under evaluation, the alternative is to generate a new cross section library. This can be accomplished with the methodology and codes in the MOCUP system. The actual mechanics of the MOCUP system are described in more detail in Reference 7. The following discussion is a brief presentation of the MOCUP code system interjected with specifics pertinent to the burnup calculations herein.

3.1 Basic Methodology

The MOCUP process begins basically with the development of an MCNP model of the reactor core. If possible, a symmetric, fractional core model or cluster of fuel elements are modelled in place of the full core in order to save model development time and computer runtime and yet still retain the spatial and spectral effects of the full core. A powerful feature of MCNP Monte Carlo method is its capability to calculate fluxes and reaction rates in very specific fuel element locations and capture the important core physics effects in the cross sections. This circumvents the more tedious and time-consuming process of using multigroup physics codes in a multi-step space and energy weighting procedure followed by a flux collapse to arrive at the same desired cross sections. The MOCUP methodology not only yields accurate results, but is also an expeditious and therefore cost-saving calculational technique relative to other methods.

The MCNP model cell location in which new cross sections are desired is tallied for the total cell flux and the fission, capture, (n,2n), and (n,3n) reaction rates for each nuclide. MCNP neutron cross section libraries are now available for calculation of reaction rates for up to 38 different actinides and 85 fission products in a given fuel composition cell. Table 3 lists these actinides and fission products. Upon completion of the calculation, MCNP writes an output file and a tally file that can be read as input by MOCUP (mcnpPRO module).

The mcnpPRO module (MCNP Post-processor) in MOCUP actually reads from five different input sources including the MCNP input, output, and tally files, as well as two special user supplied files referred to as the Nuclide Correspondence and Flux Multiplier tables and generates an mpo.a file (MOCUP Processor Output file). The mpo.a file contains a list of every tallied nuclide and the mcnpPRO calculated reaction cross sections.

The origenPRO module (ORIGEN Pre-processor) then reads nuclide and cross section data on the mpo.a file and writes it to an ORIGEN input file, thereby updating or replacing previous input file cross sections that will be substituted into the selected standard ORIGEN decay and cross section library. ORIGEN2 performs the burnup calculation over the specified time step or interval for a given flux or power level based on the LWBR power history, and outputs the new nuclide concentrations to an output file. The first burnup step or interval is now complete.

If further burnup time steps are required, the compPRO module (Composition Processor) reads the ORIGEN2 composition output files, converts these to new MCNP nuclide number densities, and creates a new MCNP input file for the next time step and the process starts over. The MOCUP code recursively continues this pattern of calculating new cross sections (updates) and number densities for each sequential time interval or burnup increment until the desired burnup or EOL is reached.

The time interval between cross section updates affects the accuracy of the burnup calculation. As the fuel burnup increases, the spatial and spectral changes influence the magnitude of the cross sections. The effect of cross section updates on accuracy is explored in the validation analysis here.

The Monte Carlo calculated results predicted by MCNP are statistical in nature and invariably raise questions concerning the error in the effective one-group cross sections and the propagation of these errors through the burnup calculation. The statistical error in the calculated total flux and reaction rates is reduced to an insignificantly small value. This was achieved by running large numbers of particles through the MCNP core models. Typically, up to 1.5 to 2.0 million initial particles are started and tracked in the calculation and result in errors significantly less than 0.5% (one standard deviation) for the fission and radiative capture cross sections of the major fuel actinides and poisons. In addition, propagation of these errors tend to be self-correcting (Reference 12) over the course of the burnup calculation. For example, underestimation of a reaction rate and hence the cross section will result in under-depletion of that nuclide for that time interval. The under-depleted isotopic concentration will in turn produce a reaction rate higher than

the true value in the next burnup interval. The higher reaction rate will produce a larger cross section causing acceleration of the depletion of the nuclide in the following interval and so on. This results in a statistical oscillation of the cross sections, reaction rates, and the nuclide concentrations as a function of burnup; the oscillation being always about the zero-error true value. Therefore, with sufficiently small MCNP statistical errors in the calculated values and the self-correcting nature of the burnup calculation in general, the effects of Monte Carlo statistics is thus reduced to a minor calculational impact.

Table 3. Actinide and Fission Product List for which Neutron Cross Section Updates are generated in the Calculations.

NUCLIDE CATEGORY	NUCLIDES
Actinides	Th-232, Th-233 Pa-233 U-232, U-233, U-234, U-235, U-236, U-237, U-238, U-239, U-240 Np-235, Np-236, Np-237, Np-238 Pu-237, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Pu-243 Am-241, Am-242m, Am-243 Cm-242, Cm-243, Cm-244, Cm-245, Cm-246, Cm-247, Cm-248 Bk-249 Cf-249, Cf-250, Cf-251, Cf-252
Fission Products	Kr-82, Kr-83, Kr-84, Kr-86 Y-89 Zr-90, Zr-91, Zr-92, Zr-93, Zr-94, Zr-95, Zr-96 Nb-93 Mo-95, Mo-97, Mo-98, Mo-99, Mo-100 Tc-99 Ru-101, Ru-102, Ru-103 Rh-103, Rh-105 Pd-105, Pd-108 Ag-107, Ag-109 Cd-113 In-115 I-127, I-129, I-135 Xe-128, Xe-129, Xe-130, Xe-131, Xe-132, Xe-133, Xe-134, Xe-135, Xe-136 Cs-133, Cs-134, Cs-135 Ba-138, Ba-140 La-139, La-140 Ce-141, Ce-142, Ce-143, Ce-144 Pr-141, Pr-143 Nd-143, Nd-145, Nd-147, Nd-148 Pm-147, Pm-148, Pm-148m, Pm-149, Pm-151 Sm-147, Sm-148, Sm-149, Sm-150, Sm-151, Sm-152, Sm-153, Sm-154 Eu-151, Eu-153, Eu-154, Eu-155, Eu-156 Gd-155, Gd-156, Gd-157, Gd-158, Gd-160 Ho-165 Er-166, Er-167

3.2 MCNP Models

Two different MCNP computer models were developed, one for the seed and one for the standard blanket and reflector fuel rods. The MCNP models are necessary for the neutron transport analysis and cross section development described in section 3.1. The following sections describe in more detail the geometry, materials, and assumptions for the two models.

3.2.1 Unit Cell Lattice Model Description (Rod N)

A single hexagonal unit cell model with reflective boundary conditions was developed to represent the neutronic conditions of a seed module fuel rod, namely, Rod N. Reflective boundaries were then applied to the unit cell hexagonal surfaces and in order to neutronically represent an infinite lattice of identical hexagonal unit cells (see Figure 8).

The infinite lattice model representation was chosen to evaluate Rod N, because Rod N is located near the center of the seed module containing 11 hexagonal rings of identical fuel rods. From the perspective of Rod N, the rest of the universe appears to be an infinite sea of identical unit cells. Each unit cell fuel rod is the same length with a central 84" high zone and a 10" thorium reflector on both the top and bottom. Rod N is essentially in the middle of a lattice of uniform fuel rods and virtually immune to the spectral and leakage effects from the peripheral low zone seed fuel rods and the surrounding standard blanket fuel rods beyond.

The hexagonal unit cell is modelled explicitly in full 3D detail. This includes exact modelling of the fuel rod diameter (0.306"), pellet diameter (0.252"), gas gap thickness (0.005"), clad thickness (0.022"), and lattice pitch (0.370"). In addition, the core operating conditions were also modelled as closely as possible to the typical steady-state LWBR operating conditions. This included use of light water at an average bulk coolant temperature of 531 F and a pressure of 2000 psi. Zircaloy-4 cladding material was used along with the 98% enriched U-233 binary oxide ($\text{UO}_2\text{-ThO}_2$) pellet material and the appropriate fissile enrichment/loadings given in Figure 2.

Figure 8 shows an x-y cross sectional view (MCNP plot) of a unit cell and the surrounding infinite lattice of unit cells. The high zone seed fuel pellet region is the green circle in the center of the unit cell, the gas gap is represented by the white annulus around the fuel pellet, the cladding by the black annulus, and the light water coolant by the blue color.

3.2.2 Standard Blanket/Seed Model Description (Rod I)

The MCNP neutron transport model developed for the standard blanket fuel rod (Rod I) is considerably more complicated than the simple unit cell modelled used in the analysis of the seed

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Figure 8. Infinite unit cell lattice model representation (MCNP) of the LWBR seed module fuel rods.

fuel rod (Rod N) analysis. The standard blanket/seed model includes in fully explicit 3D detail of both a seed module and a standard blanket module. Reflective boundary conditions applied to the six hexagonal peripheral boundaries of the standard blanket then represent, from a neutronic standpoint, the equivalent of an infinite lattice of seed and standard blanket modules. The model is shown in x-y cross section in Figure 9 (approximately at the axial midplane).

The complexity of the standard blanket/seed model was required in order to more accurately account for the complex neutron spectral and leakage effects that occur in the standard blanket regions. The various colored circles in Figure 9 allude to the wide variety of different fuel rods in the seed and standard blanket modules. The smaller diameter pink and yellow circles are high and low zone seed fuel rods, respectively. Beyond the yellow low zone seed fuel rods is a thick gray line that represents the Zircaloy seed module side or flow plate and beyond this is a blue water gap that separates the seed and standard blanket modules. The green, gray, and orange circles represent the standard blanket fuel rods in the low, medium, high, medium fuel zones, respectively.

Figure 6 shows the axial loading patterns in both the seed and standard blanket modules. Although Figure 6 is not an MCNP model output plot, all of the axial fuel rod loading details in this figure are fully incorporated into the MCNP model. In addition, the top and bottom thoria reflector regions above the active core height are also included along with the thoria steps at the center bottom of the standard blanket module.

As in the seed unit cell model for Rod N described above, the core operating conditions, geometry, materials are all modelled as closely as possible to normal steady-state LWBR operating conditions. The standard blanket lattice pitch is 0.630", the fuel pellet diameter is 0.515", clad thickness is 0.028", and the fuel rod diameter is 0.571". The seed dimensions are the same as those described in section 3.2.1. The bulk coolant temperature and pressure are 531 F and 2000 psi, respectively. Zircaloy-4 cladding material was used along with the 98% enriched U-233 binary oxide ($\text{UO}_2\text{-ThO}_2$) pellet material and the appropriate fissile enrichment/loadings given in Figure 3 for the standard blanket.

3.2.3 Reflector Fuel Rod Model Description (Rod H)

The neutron cross sections for the thoria reflector rod (Rod H) were calculated using the same model as the standard blanket fuel rod described in section 3.2.2. The cross sections were derived from the thoria top and bottom axial reflector regions (above and below the 84" active zone region). The neutron cross sections are volume-averaged over the two thoria regions. Although

Figure 9. Infinite lattice model representation (MCNP) of the LWBR standard blanket/seed modules.

an MCNP model of a reflector module (type IV) was not explicitly developed for Rod H, the thorium top and bottom regions of the fuel rods in the seed and standard blanket sections should produce reasonable spectrally and spatially flux-weighted neutron cross sections relative to those that could have been obtained in an actual reflector module model.

3.3 Cross Section Updates

In order to get an accurate estimate of the radionuclide inventory, characteristic neutron reaction cross sections are an important parameter for the depletion calculation. Neutron cross sections can vary significantly over a given burnup period and need to be corrected or updated appropriately at certain time intervals or steps. The greater the number of cross section updates the better the depletion calculation. Of course, there is a point in which the calculation will begin to converge (4 to 8 updates for the LWBR) and additional updates will not significantly improve the calculated values.

Of primary importance are the Beginning-of-Life (BOL) cross sections, i.e. the first cross section update and the first set of cross sections that go into the depletion calculation. The development and usage of these cross sections have a greater impact on the depletion calculation than do subsequent cross section updates. The BOL cross sections are exact for the start of the depletion calculation and are specific to a reactor type or even a particular position in the reactor core. It is important from an accuracy standpoint, that reactor specific BOL cross sections be used as opposed to a readily available standard cross section library for a different reactor.

Of secondary importance to the BOL cross sections are cross section updates as a function of burnup. Although the cross section updates as a function of burnup are important (usually 4 to 8 updates are sufficient to achieve reasonable convergence for a depletion calculation), the development and usage of the BOL cross sections outweigh the effects of these updates.

The BOL and update cross sections are highly-dependent on the local neutron spectrum which in turn is governed by such basic reactor features such as the moderator or coolant (e.g. light water, heavy water, beryllium- or graphite-moderated, sodium-cooled, etc.), enrichment, lattice pitch, local absorbers (targets, breeding material, control rods, parasitic absorbers, etc.), reflectors, uranium loadings, proximity to different loaded or burned assemblies, etc. The number of variables and their synergistic interactions can be quite complex, hence the usage of a three-dimensional transport code (MCNP) and inclusion of as many core geometry features as possible in the calculations.

3.3.1 Rod N Updates

Seed fuel rod cross sections are developed based on the unit cell infinite lattice model described in section 3.2.1. Cross sections are developed for 1, 2, 4, and 8 updates over the burnup or operating period. The first update is of course the BOL cross sections. In the results section, the effect of cross section updates on the isotopic concentrations is given.

3.3.2 Rod I Updates

Standard blanket fuel rod cross sections are developed based on the model described in section 3.2.2. Cross sections are developed for 1, 2, and 4, updates over the burnup or operating period. The first update is again the BOL cross sections. In the results section, the effect of cross section updates on the radionuclide concentrations is given.

3.3.3 Rod H Updates

Reflector fuel rod cross sections are developed based on the model described in sections 3.2.2 and 3.2.3. Cross sections are developed for only 1-update over the burnup or operating period. These cross sections represent just the BOL cross sections in the thoria reflector regions. Concentrations of other radionuclides including U-233 are relatively small even at EOL and impact the neutron cross sections very little as a function of burnup.

4.0 EXPERIMENTAL DATA

Section 4.1 begins with a brief description of the destructive chemical assay process used by Argonne National Laboratory to determine the fissile and fission product concentrations in the 17 assayed LWBR fuel rods. The process description is based on process descriptions from References 5 and 13.

Sections 4.2, 4.3, and 4.4 give a brief technical description of the assayed fuel rods N, I, and H, respectively, along with the destructive assay EOL experimental uranium isotopic data. The EOL uranium isotopic data are taken directly from the assay reports documenting the chemical and radiometric analyses performed at the Argonne National Laboratory under the LWBR-POB Analytical Support Project (References 2, 3, and 4). A complete listing of all the reports and test results can be found in Reference 5. Experimental assay data from three of these reports were used to compare with the calculated results.

4.1 Destructive Assay Process

Thirty-three LWBR EOL fuel rods were selected from a 500-rod POB (Proof-of-Breeding) sample pool for destructive chemical assay. Seventeen of these rods were then subjected to extensive chemical and physical measurements. Measurements included the determination of the mass and abundance of the uranium isotopes, the total mass of uranium, moles of Cs-137, Ce-144, Zr-95, xenon isotopes, and krypton isotopes, in addition to rod and segment weight, length, and oxide fuel and clad masses. The experiment errors associated with these measurements are also reported.

Initially, each whole fuel rod was weighed, its full rod length measured, and an axial gamma-scan and a non-destructive assay by the PIFAG (Production Irradiated Fuel Assay Gage) were performed. The fuel rods were then sheared into pre-defined lengths or contiguous segments and collected in aluminum buckets. The segment lengths ranged from 10.5" to 14" in length, and the weight and length of each segment was measured. The shearing also served to pulverize the ceramic thorium-uranium fuel material to facilitate its dissolution. The segments (and bucket) were then dissolved in one of two high pressure, high temperature dissolver systems. Also, the fuel rod end pieces were weighed and any particles of fuel material in the rod plenum were recovered for analysis, so that the total fissile uranium content could be accounted for.

The dissolution of the individual sheared segments (cladding plus fuel) was carried out using a 4-hour dissolution in Thorex (13.6 M HNO₃, 0.06 M HF) followed by a dilute nitric acid rinse, then

a 3-hour secondary dissolution in a Thorex-0.06 M Al^{+3} solution, followed by a reflux rinse (hot rinse) and cold rinse with dilute nitric acid. The segments were subjected to the fluoride-catalyzed nitric acid, boiling under reflux at elevated temperature (195 C) and pressure (120 psig), in order to dissolve the pulverized fuel material. A sample of the second dissolution was obtained to measure the completeness of the dissolution scheme. Gases emitted during the dissolution were collected and analyzed for Kr and Xe.

Both dissolution solutions from each segment were then transferred, along with rinses, from the dissolver into a blend tank where the solution was weighed and mixed. From the mixed solution, small samples were taken for isotopic uranium analysis and for total uranium analysis by mass spectrometric isotopic dilution (MSID), employing NBS SRM 993 as the reference spike (uranium-235). A weighed amount of NBS SRM 950a was dissolved as a reference spike (uranium-238) in the remaining major portion of the solution in the blend tank, and additional samples of the solution were taken for total uranium analysis by MSID and for radiochemical determination of the fission product monitors cesium-137, cerium-144, and zirconium-95. This fissile uranium assay was based on the MSID analysis using the U-238 spike. The results with the U-235 spike are used as corroborative data.

In all, three sets of samples were obtained from each segment. The first set (two samples) was taken from the secondary dissolution prior to blending to assess the completeness of the dissolution scheme. The second set (four samples) was then taken from the blended (both dissolutions and all rinses) tank contents. The third set (four samples) was taken after the addition of the known amount of U-238 spike (NBS Standard Sample 950a). Half of the samples were analyzed, the other half were placed in archive. Batch carry over or cross contamination was controlled and monitored by analyzing a blank (a full dissolution scheme with no segment material) between each rod and all segments were analyzed in order of increasing U content.

Implicit in all measurements is that dissolution is complete and fuel in solution is quantitatively transferred to the blend tank.

The uranium analyses included the measurement of the total uranium mass and uranium isotopic mass and abundance (U-233, U-234, U-235, U-236, U-238) by thermal ionization mass spectrometry. Due to the interference of Th-232, the U-232 isotopic mass was alternatively determined by alpha spectroscopy.

Fission gases (Kr and Xe) collected from the fuel rod plenum and during dissolution were determined by gas mass spectrometry on a "best effort" basis. Gases released during shearing were estimated by using the in-line radiation monitor in the cell ventilation system. The plenum (rod void volume) contained between 0.01% and 0.15% of the total Kr + Xe gas, an additional 0.17% to 0.58% of the total Kr + Xe gas was released during shearing, and the rest (>99%) was released during the dissolution process.

The fission products, Cs-137, Ce-144, and Nb-95 (Zr-95 decay daughter) were determined by gamma spectrometry (high purity germanium detector with associated automated multi-channel analyzer/data management system) on weighed aliquots of the samples obtained prior to spiking the blend tank with 950a (U-238). Cs-137 and Ce-144 were determined on a sample aliquot by direct counting. Zr-95 was obtained after processing the sample aliquot through a cleanup procedure to reduce interferences. The losses of Zr-95 were accounted for by using before and after values of the Ce-144. Error requirements for Zr-95 measurements made after 10/84 were waived, due to the short half-life (64.02 days).

The ANL destructive assay program assayed 33 fuel rods from seed, standard blanket, power-flattening blankets, and reflector modules. The total number of rod segments dissolved was 298, the weight of $\text{UO}_2\text{-ThO}_2$ dissolved was 105.5 kg, and 298 determinations of U-total, U-isotopic, and fission products (Cs-137, Ce-144, and Zr-95). The objective of the program was twofold: (1) provide accurate and precise total fissile uranium loading in each of the 33 POB sample rods and (2) provide analytical results to permit correction of self-shielding effects on the PIFAG measurements.

4.2 Rod N Assay Data

The first fuel rod selected for a validation comparison between calculated and assay radionuclides was seed module Rod N. Rod N was sheared into 10 segments and the axial midplane segment 3 was further chosen as the segment isotopics that would ultimately be used in the comparison. Rod N (segment 3) is a high zone, binary oxide ($\text{UO}_2\text{-ThO}_2$) pellet fuel rod located near the very center of the seed module at core midplane. These center seed fuel rods experienced some of the highest burnups in the LWBR core (approximately 30 GWd/MTHM average to 54 GWd/MTHM maximum).

The exact location of fuel rod N in a seed module is shown in Figure 2. Rod N occupied the seed module location C-10. The "C" refers to the third hexagonal ring from the center of the module. The hexagonal rings of the module are alphabetically labelled from "A" (center of the module) to

“Q” (periphery of the seed module). In addition, the C-10 designator further indicates that Rod N occupied the tenth position in the “C” or third hex ring. The positions in a ring are assigned numerical values from 1 to N (N varies from ring to ring) in a clockwise manner starting from the upper right 45-degree diagonal (see Figure 2). The specific rod identification number for Rod N was 0507057.

The destructive assay of Rod N began by segmenting (or shearing) the length of the rod into 10 contiguous segments. Segment 3 contained the geometric axial center of the 84” high zone of the fuel rod. The length of segment 3 was 21.171” with a weight of 210.831 grams (including the clad). At BOL, pellets in this segment would have had a diameter of 0.252” and a corresponding volume of 17.30 cm³.

The destructive assay (fuel dissolution, and chemical and radiometric analyses) of this rod took place over the period of June 27 to July 29, 1985. The midpoint of this period (July 16, 1985) was the decay date chosen to evaluate the actinide and fission product concentrations for comparison with the chemical and radiometric analyses.

Table 4 below gives the EOL uranium isotopic percent abundances and masses which were measured from the chemical and radiometric analyses performed on Rod N (segment 3):

Table 4. Assayed uranium isotopics for Rod N segment 3.

Isotope	Weight Percent (wt %)	Segment Mass (grams)	Experimental Error (+/-) 1-sigma
U-232	0.1181	0.007754	0.000243
U-233	86.2675	5.661795	0.001350
U-234	11.1477	0.731631	0.000185
U-235	2.0017	0.131371	0.000264
U-236	0.1621	0.010636	0.000007
U-238	0.3029	0.019878	0.000256

The uranium EOL isotopic assay data was taken directly from Reference 2. Other fission product radionuclide concentrations are also reported in this reference and will be presented and discussed in following sections.

Rod N (segment 3) was selected to be compared with the depletion calculation for three reasons: (1) the rod is centrally located in a virtual infinite array of similar high zone fuel rods, thereby, minimizing the effects of neutron leakage and spectra effects from the surrounding standard blanket, (2) segment 3 is in the very center of the fuel rod, thereby, minimizing the effects of the top and bottom thoria reflectors, and (3) from Reference 14, estimates of burnup (peak and average) in MWd/MTHM were available for another seed fuel rod (B-4) in the immediate vicinity of Rod N and therefore, Rod N would be expected to have a similar burnup. These referenced burnups of other rods in the vicinity of the rods under evaluation proved to be very valuable for ensuring that calculated burnup estimates were on target.

4.3 Rod I Assay Data

The second fuel rod selected for a validation comparison between calculated and assay radionuclides was standard blanket module Rod I. Rod I was a high zone, binary oxide ($\text{UO}_2\text{-ThO}_2$) pellet region located near the periphery of a standard blanket module at location E-56 (see Figure 3). These blanket fuel rods experienced the medium to low burnups in the LWBR core (5-23 GWd/MTHM). The exact location of Rod I in the standard blanket module is shown in Figure 3. The specific rod identification number is Rod No. 0507057.

The destructive assay of this rod took place over the period of February 4 to March 11, 1985. The midpoint of this period (February 23, 1985) was the decay date chosen to evaluate the actinide and fission product concentrations for comparison with the chemical and radiometric analyses.

The destructive assay of Rod I also began by segmenting (or shearing) the length of the rod into 10 contiguous segments. Segment 2 was chosen for the calculational evaluation and was the top segment of the 84" high zone fuel region just below the top thoria axial reflector region. The segment 2 length was 14.297" and had a weight of 536.941 grams. At BOL, pellets in this segment would have had a diameter of 0.572" and a corresponding volume of 47.95 cm³.

Table 5 below gives the EOL uranium isotopic percent abundances and masses that were measured from the chemical and radiometric analyses performed on Rod I (segment 2). The uranium EOL isotopic assay data was taken directly from Reference 3. Other fission product radionuclide concentrations are also reported in this reference and will be presented and discussed in following sections.

Table 5. Assayed uranium isotopics for Rod I segment 2.

Isotope	Weight Percent (wt %)	Segment Mass (grams)	Experimental Error (+/-) 1-sigma
U-232	0.0358	0.003328	0.003330
U-233	94.0117	8.735407	0.002300
U-234	5.1233	0.476052	0.000214
U-235	0.5588	0.051922	0.000409
U-236	0.0370	0.003434	0.0000186
U-238	0.2334	0.021689	0.000400

Rod I (segment 2) was selected to be compared with the depletion calculation for two reasons: (1) it is near the top of the fuel rod and hence the axial reflector variation effects are minimized, and (2) from Reference 14 estimates of burnup (peak and average) in MWd/MTHM were available for an adjacent fuel rod in location E-57.

4.4 Rod H Assay Data

The third fuel rod selected for a validation comparison between calculated and assay radionuclides was reflector module Rod H. Rod H is from a Type IV reflector module. Its BOL fuel composition was pure thoria oxide ($\text{UO}_2\text{-ThO}_2$) with no urania present. Rod H was located near the apex of the reflector module IV in the first row (position B1) adjacent to the power-flattening blanket region. See Figure 5 for the exact location of Rod H with the reflector module and Figure 7 relative to the LWBR core. Reflector rods closest to the core received the highest burnups (greatest irradiation flux from the core and hence maximum breeding) relative to other reflector rods further from the core and closer to the pressure vessel boundary. Consequently, high burnup and breeding reflector rods, such as Rod H possessed the highest concentrations of bred U-233 relative to other lower burnup reflector rods. Maximum reflector rod burnups are estimated to be approximately 4,100 MWd/MTHM (Reference 14). The burnup for Rod H was calculated to be approximately 3,515 MWd/MTHM, i.e. in the end of the burnup high range. The specific rod identification number is Rod No. 0507057.

The destructive assay of this rod took place over the period of January 3 to February 26, 1985. The midpoint of this period (January 31, 1985) was the decay date chosen to evaluate the actinide and fission product concentrations for comparison with the chemical and radiometric analyses.

The destructive assay of Rod H also began by segmenting (or shearing) the length of the rod into 15 contiguous segments. Segment 7 was chosen for the calculational evaluation because it contained the mid-section of the thoria reflector rod or the geometric axial midpoint of the rod where one might expect the highest burnup to occur. The segment 7 length was 6.982" and had a weight of 556.021 grams. At BOL, pellets in this segment would have had a diameter of 0.832" and a corresponding volume of 49.41 cm³.

Table 6 below gives the EOL uranium isotopic percent abundances and masses which were measured from the chemical and radiometric analyses performed on Rod H (segment 7):

Table 6. Assayed uranium isotopics for Rod H segment 7.

Isotope	Weight Percent (wt %)	Segment Mass (grams)	Experimental Error (+/-) 1-sigma
U-232	0.1259	0.004893	0.000152
U-233	94.2144	3.661613	0.000791
U-234	5.0898	0.197812	0.000050
U-235	0.5438	0.021134	0.000222
U-236	0.0214	0.000833	0.00000389
U-238	0.0048	0.000185	0.000194

The uranium EOL isotopic assay data was taken directly from Reference 4. Other fission product radionuclide concentrations are also reported in this reference and will be presented and discussed in following sections.

Rod H (segment 7) was selected to be compared with the depletion calculation for two reasons: (1) the rod was in the first row of the module reflector rods closest to the power-flattening blanket and hence would have experienced one of the highest possible burnups of any of the reflector module rods, and therefore, also would have one of the highest reflector rod EOL U-233 concentrations due to breeding, and (2) from Reference 14 an estimate of peak reflector rod burnup of 4,100 MWd/MTHM was available.

4.5 Other Module Fuel Rods and Segments

From the 17 LWBR fuel rods chemically assayed, there are approximately 270 other fuel rod segments that could have been analyzed in the same fashion as Rod N (segment 3), Rod I (segment 2), and Rod H (segment 7). Additional rod segments or segments from other modules besides the

seed, standard blanket, and reflector modules could also be evaluated. Of course, calculating more would be better, but the author believes that the outcome would be redundant results with little additional insight gained from the additional effort of modelling. These three evaluations and comparisons represent a reasonable sample and provide sufficient sampling of the segments to demonstrate the calculation accuracy and trends relative to the measured data. The other experimental data is certainly available for additional validation, if needed.

5.0 RESULTS COMPARISON

The following three sections (5.1, 5.2, 5.3) present calculated EOL uranium and fission product isotopic mass concentrations and compare them with measured data for the three different fuel rod segments, i.e. Rod N segment 3, Rod I segment 2, and Rod H segment 7. The calculated uranium weight percentages at EOL are also compared with the measured data. In addition, for Rod N and Rod I, the effect on the calculational accuracy of increasing the number of neutron cross section updates as a function of burnup is also provided.

The comparison tables showing the calculated versus measured values also contain the BOL uranium loading estimates for each segment evaluated. These BOL uranium segment mass loadings are based on the BOL fuel pellet diameters, the BOL calculated number densities, and the EOL sheared segment length recorded for each assayed fuel rod.

The segment burnups are based on LWBR power history and normalized to the measured EOL assay concentration of Cs-137. The Cs-137 concentration was probably measured specifically for this purpose, since estimated segment burnups were not supplied with the chemical assay data.

5.1 Rod N Segment 3 Results Comparison

For the seed fuel rod (Rod N), the calculated EOL uranium and fission product isotopic mass concentrations are compared with the measured assay EOL data in Tables 7, 8, and 9. Calculated data in Tables 7 and 8 are based on a burnup of 37,000 MWd/MTHM and 8 cross section updates, i.e. these calculated values represent our “best” estimate. Table 9 shows the effect on the calculational accuracy for increasing the number of cross section updates.

Table 7 presents a comparison of the BOL, measured EOL, and calculated EOL uranium, krypton, xenon, cesium-137, cerium-144, and zirconium-95 isotopic masses for segment 3 of Rod N. The far column labelled “DIFF” is the difference between the calculated and measured values and is defined as: $((\text{Calc}-\text{Assay})/\text{Assay}) \times 100$. With this definition, a negative difference value indicates that the calculation under predicted the measured assay value, and a positive difference value indicates that the calculation overpredicted the measured assay value. Also included in a column to the right of the “ASSAY EOL” (or assay measurement at EOL) are the measurement errors associated with the measured mass values.

Table 7. Comparison of measured versus calculated uranium and fission product mass isotopics for a maximum of 8 cross section updates and 37,000 MWd/MTHM burnup (Rod N).

ISOTOPE	BOL (g)	ASSAY EOL (g)	Experimental Error (g)	CALC EOL (g)	DIFF* EOL (%)
U-232	0.000000	0.007754	0.000243	0.010920	40.8
U-233	8.959900	5.661800	0.001351	5.747000	1.5
U-234	0.117700	0.731630	0.000185	0.764300	4.5
U-235	0.008210	0.131370	0.000264	0.148000	12.7
U-236	0.001824	0.010636	0.000007	0.013360	25.6
U-238	0.033767	0.019880	0.000256	0.026810	34.9
Kr-82	0.000000	0.000216	0.000030	0.000349	61.8
Kr-83	0.000000	0.017147	0.002379	0.018630	8.6
Kr-84	0.000000	0.033048	0.004568	0.040000	21.0
Kr-85	0.000000	0.006375	0.000888	0.007225	13.3
Kr-86	0.000000	0.054883	0.007584	0.063960	16.5
Xe-128	0.000000	0.000608	0.000093	0.001967	223.3
Xe-130	0.000000	0.000618	0.000095	0.003617	485.3
Xe-131	0.000000	0.073480	0.011263	0.076570	4.2
Xe-132	0.000000	0.150590	0.023055	0.194700	29.3
Xe-134	0.000000	0.169440	0.025939	0.203900	20.3
Xe-136	0.000000	0.242440	0.037126	0.293600	21.1
Cs-137	0.000000	0.202650	0.000561	0.200900	-0.9
Ce-144	0.000000	0.008404	0.000057	0.002982	-64.5
Zr-95	0.000000	0.000032	0.000006	1.165E-07	-99.6

* End-of-Life Difference = DIFF = ((Calc-Assay)/Assay) * 100

Table 8. Comparison of measured versus calculated uranium mass isotopic percentages for a maximum of 8 cross section updates and 37,000 MWd/MTHM burnup (Rod N).

ISOTOPE	BOL (g)	ASSAY EOL (g)	CALC EOL (g)	DIFF* EOL (%)
U-232	0.00	0.12	0.16	37.8
U-233	98.23	86.27	85.64	-0.7
U-234	1.29	11.15	11.39	2.2
U-235	0.09	2.00	2.21	10.2
U-236	0.02	0.16	0.20	22.8
U-238	0.37	0.30	0.40	31.9

* End-of-Life Difference = DIFF = ((Calc-Assay)/Assay) * 100

Again, because the burnup estimates for each segment were not provided with measured isotopic mass values, the segment burnup was iteratively derived from the calculation by matching the calculated Cs-137 mass concentration to the measured Cs-137 mass concentration. In Table 7, the calculated Cs-137 mass value is very close to the measured Cs-137 mass value (i.e. a difference of only -0.9%). This resulted in a burnup estimate of approximately 37,000 MWd/MTHM for segment 3 of Rod N. A further iteration reveals a more refined estimated burnup value of 37,330 MWd/MTHM.

Using the 37,000 MWd/MTHM burnup estimate, there appears to be good agreement between the measured and calculated **uranium** isotopic masses. The U-233 mass difference is only +1.5%, which is a good indication that the 8 cross section updates were sufficient to accurately capture the complicated neutronic production and destruction of the U-233 mass as a function of burnup. The next significant uranium mass isotopes are U-234 and U-235 with differences of +4.5% and +12.7%, respectively. The other uranium isotopes are of relatively low mass concentrations, but remain within a +41% difference. Note all the calculated uranium isotopic masses are conservatively predicted relative to the measured masses.

The calculated U-236 concentration exhibits only a +5.0% difference which is in very good agreement. The U-236 isotope is produced primarily from radiative capture by U-235 and destroyed by radiative capture to U-237. Since the U-235 concentration are relatively small to begin with, the U-236 concentration is even smaller.

The calculated U-232 concentration exhibits a +40.8% difference which is reasonable agreement considering the isotope's relatively more complex production and destruction chains. The production of U-232 appears to have at least two major potential pathways or chains: (1) radiative capture by Th-232 to Th-233, Th-233 beta-decay to Pa-233, Pa-233 beta-decay to U-233, and then an (n,2n) reaction with U-233 to create U-232, and (2) an (n,2n) reaction with Th-232 to Th-231, beta-decay of Th-231 to Pa-231, radiative capture by Pa-231 to Pa-232, and finally a beta-decay of Pa-232 to U-232. U-232 can then undergo destructive processes, i.e. radiative capture, (n,2n), etc. reactions and alpha decay. Therefore, the production of U-232 is relatively complicated and dependent on the many reaction cross sections which in turn makes it more difficult to accurately calculate.

The calculated U-238 concentration exhibits a +34.9% difference which again is reasonable agreement. The calculated U-238 concentration is dependent on the initial BOL concentration in

the oxide fuel meat and its radiative capture cross section. The initial BOL concentration would in turn be sensitive to the accuracy of the BOL uranium isotopic enrichment weight percentages.

The **krypton** isotopes (Kr-83, Kr-84, Kr-85, Kr-86) have differences of less than or equal to 21%, with the exception of the very low yield and low mass, Kr-82 isotope (62% difference). The Kr-82 isotope is a “shielded” fission product (Se-82) and is presumed to have a direct, but exceedingly small fission yield of less than $2.5\text{E-}6\%$. Kr-82 is probably produced primarily from the beta-decay of Br-82 (another “shielded” fission product) also with an apparently small but more significant direct fission yield. Another production path for Kr-82 may also be the result of radiative capture and high energy threshold reactions with both impurity nuclides in the oxide fuel and/or fission product nuclides. In addition, note the sizable experimental errors associated with the krypton isotopic mass measurements.

The stable isobar fission product **xenon** isotopes (Xe-131, Xe-132, Xe-134, Xe-136), whose production is driven primarily from multiple beta-decays of neutron-rich fission products, show a difference of $<30\%$. The other two xenon isotopes measured (Xe-128, Xe-130) show very poor agreement with differences of 96.8% and +485.3%, respectively. Although both isotopes are stable, they are “shielded” fission products whose direct fission yields are presumed to be less than $2.5\text{E-}6\%$. Production of these two xenon isotopes is (in addition to the minute direct fission yields) most probably due to a complex combination of radiative captures (n,γ) and high energy threshold particle reactions (e.g. $(n,2n)$, $(n,3n)$, (n,p) , (n,α) , etc) with neighboring nuclides. The neighboring nuclide concentrations are affected by both initial oxide fuel impurities (not considered in the calculations here) and the result of direct fission events. Without the inclusion of the oxide impurities and modelling of only some of the reactions (i.e. (n,γ) , $(n,2n)$, $(n,3n)$) that affect their concentration, accurate calculation these two low mass xenon isotopes was not expected.

However, reasonable agreement was obtained for the Xe-128 and Xe-130 isotopic masses for Rod I below. Hence, it is possible that the measured values are not very accurate. Note also the sizable experimental errors associated with the xenon isotopic mass measurements.

The **cesium** isotope (Cs-137) mass value reported in Table 7 shows a difference of only -0.9%. This difference is very small because it was used to “match” the calculation to measurement, and hence a zero difference with successive iterations in the approach to an exact matching would be expected. The Cs-137 isotope was chosen to match the calculation and measurement values for the reasons given in section 1.0 and at the same time provided what the author believes is a very

reasonable estimate of the burnup. The measured value has a very small experimental error and confidence in this measured value is expected to be relatively high.

The **cerium** isotope (Ce-144) shows a difference of -64.5%. The relatively large difference and the fact that the difference is negative indicates that the calculation underpredicts the measured concentration. The Ce-144 isotope is radioactive with a short half-life (284.6 days). If indeed, the measurement value is accurate, potential problems in the calculation could be the result of poor fission product yields or a radiative capture cross section that is too large, or both.

The **zirconium** isotope (Zr-95) shows almost a 100% difference and in this case, the calculated value is almost negligible compared with the measured mass. After the calculations were complete, it was determined that the zirconium clad had been dissolved along with the oxide fuel pellet. The irradiation of the cladding would have produced significant quantities of Zr-95 from the transmutation of Zr-94 (natural stable zirconium isotope) present in the clad. The transmutation of Zr-94 into Zr-95 in the clad clearly would far outweigh the amount of Zr-95 produced from the fission of the uranium isotopes in the fuel pellet. Using the measured mass concentrations of Zr-95 and back-calculating, there must have been on the order of 1.02 grams of Zr-95 in the dissolved solution at the time of the isotopic measurements. This is based on the fact that approximately 15 half-lives had elapsed before the measurement took place. Hence, the large difference in the calculated and measured values. This discrepancy is the same for Rod I and H.

Table 8 shows a comparison of the measured and calculated EOL uranium isotopics by percentage of the total uranium segment mass. The percentages are in good agreement and it is concluded that the distribution of uranium isotopes is well predicted by the calculations.

Table 9 shows the effect on the calculated uranium and fission product mass isotopics as a function of the number of cross section updates over the LWBR operation cycle. The total integrated burnup for each case is still 37,000 MWd/MTHM. Of particular interest is the reduction in the difference between the measured and calculated values with the increase in the number of cross section updates. This is however not true for all the isotopes and very little improvement or change is realized between 4 and 8 updates (note: the amount of calculational effort required in going from 4 to 8 updates doubles). Our "best" estimate is obtained with the maximum number of cross section updates, but notice how well the single update or BOL cross sections predict the isotopic masses relative to the higher number of update cases. In addition, relative to the 8 update case, the differences between the calculated and measured isotopic masses for the single update are all positive or conservatively estimated with the exception of U-234 and Xe-131. However, these

Table 9. Comparison of calculated uranium and fission product mass concentrations as a function of the number of cross section updates (Rod N).
(Burnup is fixed at 37,000 MWd/MTM)

ISOTOPE	BOL (g)	ASSAY EOL (g)	1 update		2 updates		4 updates		8 updates	
			CALC EOL (g)	DIFF* EOL (%)	CALC EOL (g)	DIFF* EOL (%)	CALC EOL (g)	DIFF* EOL (%)	CALC EOL (g)	DIFF* EOL (%)
U-232	0.000000	0.007751	0.011700	50.9	0.010910	40.8	0.010830	39.7	0.010920	40.9
U-233	8.959900	5.661800	5.872000	3.7	5.825000	2.9	5.749000	1.5	5.747000	1.5
U-234	0.117700	0.731630	0.702700	-4.0	0.750300	2.6	0.761900	4.1	0.764300	4.5
U-235	0.008210	0.131370	0.197000	50.0	0.159400	21.3	0.147800	12.5	0.148000	12.7
U-236	0.001824	0.010639	0.016110	51.4	0.014370	35.1	0.013390	25.9	0.013360	25.6
U-238	0.033767	0.019880	0.026600	33.8	0.026750	34.6	0.026820	34.9	0.026810	34.9
Kr-82	0.000000	0.000216	0.000373	72.9	0.000360	66.8	0.000349	61.6	0.000349	61.8
Kr-83	0.000000	0.017147	0.018610	8.5	0.018630	8.6	0.018620	8.6	0.018630	8.6
Kr-84	0.000000	0.033048	0.040080	21.3	0.040050	21.2	0.040010	21.1	0.040000	21.0
Kr-85	0.000000	0.006375	0.007236	13.5	0.007232	13.4	0.007225	13.3	0.007225	13.3
Kr-86	0.000000	0.054883	0.064040	16.7	0.064020	16.6	0.063950	16.5	0.063960	16.5
Xe-128	0.000000	0.060844	0.002141	-96.5	0.002002	-96.7	0.001965	-96.8	0.001967	-96.8
Xe-130	0.000000	0.000618	0.003818	517.8	0.003700	498.8	0.003615	485.0	0.003617	485.3
Xe-131	0.000000	0.073480	0.068830	-6.3	0.074210	1.0	0.076520	4.1	0.076570	4.2
Xe-132	0.000000	0.150590	0.202300	34.3	0.196900	30.8	0.194800	29.4	0.194700	29.3
Xe-134	0.000000	0.169440	0.203900	20.3	0.203900	20.3	0.203900	20.3	0.203900	20.3
Xe-136	0.000000	0.242440	0.291700	20.3	0.291400	20.2	0.294600	21.5	0.293600	21.1
Cs-137	0.000000	0.202650	0.200900	-0.9	0.200900	-0.9	0.200900	-0.9	0.200900	-0.9
Ce-144	0.000000	0.008404	0.002989	-64.4	0.002984	-64.5	0.002982	-64.5	0.002982	-64.5
Zr-95	0.000000	0.000032	1.165E-07	-99.6	1.165E-07	-99.6	1.165E-07	-99.6	1.165E-07	-99.6

* End-of-Life Difference = DIFF = ((Calc-Assay)/Assay) * 100

two isotopes are only 4% and 6% underestimated relative to the measured value. This observation provides a reasonable justification for the defense of the calculated LWBR module radionuclide inventories based on BOL cross sections only.

5.2 Rod I Segment 2 Results Comparison

For the standard blanket fuel rod (Rod I), the calculated EOL uranium and fission product isotopic mass concentrations are compared with the measured assay EOL isotopic mass concentrations in Tables 10, 11, and 12.

Table 10 presents a comparison of the BOL, measured EOL assay, and calculated EOL uranium, krypton, xenon, cesium-137, cerium-144, and zirconium-95 isotopic masses for segment 2 of Rod I. The far column labelled "DIFF" is difference between the calculated and measured values and is defined as: $((\text{Calc}-\text{Assay})/\text{Assay}) \times 100$. With this definition, a negative difference value indicates that the calculation under predicted the measured assay value, and a positive difference value indicates that the calculation overpredicted the measured assay value. Also included in a column to the right of the "ASSAY EOL" (or assay measurement at EOL) are the measurement errors associated with the measured mass values. Note that only 4 cross section updates were performed for these calculations.

Again, because the burnup data were not provided along with measured isotopic mass values, the burnup assumed in the calculation was achieved when the measured mass value of Cs-137 was matched. In Table 10, the calculated Cs-137 mass value is very close to the measured Cs-137 mass value (i.e. a difference of only -1.3%). This resulted in a burnup estimate of approximately 7,000 MWd/MTHM for segment 2 of Rod I. A further iteration reveals a more refined estimate burnup value of 7,075 MWd/MTHM. This burnup level is relatively low as expected, since this segment is at the very top of the standard blanket fuel rod active zone. This region, namely, at the top of the active fuel zone and just below the top thorium reflector would have had a reduced neutron flux level relative to the lower elevation fuel segments, because core power over most of the LWBR operating cycle would have been generated near the bottom of the core where the seed modules enter and generate power.

There is good agreement between the measured and calculated uranium isotopic mass concentrations. The U-233 mass difference is only +6.2%, which is a good indication that the 4 cross section updates were probably sufficient, instead of going to 8 updates. The U-234, U-235, U-236 concentrations are quite good with +5.4, +2.7, and +5.1% differences, respectively. The

Table 10. Comparison of measured versus calculated uranium and fission product mass isotopics for 4 cross section updates and 7,000 MWd/MTHM burnup (Rod I).

ISOTOPE	BOL (g)	ASSAY EOL (g)	Experimental Error (g)	CALC EOL (g)	DIFF* EOL (%)
U-232	0.000000	0.003327	0.000102	0.004842	45.6
U-233	9.550038	8.735400	0.002301	9.109000	4.3
U-234	0.125418	0.476050	0.000214	0.499400	4.9
U-235	0.008750	0.051923	0.000409	0.053580	3.2
U-236	0.001945	0.003438	0.000019	0.003610	5.0
U-238	0.035972	0.021687	0.000400	0.033310	53.6
Kr-82	0.000000	0.000137	0.000070	0.000073	-46.7
Kr-83	0.000000	0.010654	0.001330	0.011010	3.3
Kr-84	0.000000	0.021145	0.002639	0.020710	-2.1
Kr-85	0.000000	0.004109	0.000517	0.003959	-3.7
Kr-86	0.000000	0.034838	0.004345	0.034530	-0.9
Xe-128	0.000000	0.000447	0.000020	0.000338	-24.5
Xe-130	0.000000	0.000454	0.000020	0.000683	50.3
Xe-131	0.000000	0.052169	0.002373	0.052870	1.3
Xe-132	0.000000	0.104670	0.004694	0.091110	-13.0
Xe-134	0.000000	0.120770	0.005410	0.108400	-10.2
Xe-136	0.000000	0.190040	0.008494	0.144300	-24.1
Cs-137	0.000000	0.109590	0.000484	0.108200	-1.3
Ce-144	0.000000	0.007790	0.000053	0.002270	-70.9
Zr-95	0.000000	0.000042	0.000002	2.953E-07	-99.3

* End-of-Life Difference = DIFF = ((Calc-assay)/Assay) * 100

Table 11. Comparison of measured versus calculated uranium mass isotopic percentages for 4 cross section updates and 7,000 MWd/MTHM burnup (Rod I).

ISOTOPE	BOL (%)	ASSAY EOL (%)	CALC EOL (%)	DIFF* EOL (%)
U-232	0.00	0.04	0.05	39.4
U-233	98.23	94.01	93.87	-0.1
U-234	1.29	5.12	5.15	0.5
U-235	0.09	0.56	0.55	-1.2
U-236	0.02	0.04	0.04	0.5
U-238	0.37	0.23	0.34	47.1

* End-of-Life Difference = DIFF = ((Calc-assay)/Assay) * 100

U-232 and U-238 now deviate significantly from the other uranium isotopes, but are not unreasonable at +43.8 and +56.5% differences, respectively. Note however that the larger differences for U-232 and U-238 are still positive (as are the other calculated uranium isotopic masses) and therefore would represent conservative estimates relative to the measured values.

The **krypton** isotopes (Kr-83, Kr-84, Kr-85, Kr-86) have differences of less $\pm 4\%$, with the exception of the very low yield and low mass, Kr-82 isotope (-47.5% difference). The difficulty in performing an accurate calculation for the Kr-82 isotope is believed to be for the same reasons mentioned above for the Rod N calculation.

The stable isobar fission product **xenon** isotopes (Xe-131, Xe-132, Xe-134, Xe-136), whose production is driven primarily from individual isobaric beta-decays chains of neutron-rich fission products, show a difference of $<25\%$. The other two xenon isotopes measured (Xe-128, Xe-130) show reasonable agreement with differences of -25.8% and +47.9%, respectively.

The **cesium** isotope (Cs-137) mass value reported in Table 10 shows a difference of only -1.3%. This difference is very small because it was used to "match" the calculation to measurement, and hence one would expect a zero difference with successive iterations in the approach to an exact matching. The Cs-137 isotope was chosen to match the calculation and measurement values for the reasons given in section 1.0 and at the same time provided what the author believes is a very reasonable estimate of the burnup. The measured value has a very small experimental error and confidence in this measured value is expected to be relatively high.

The **cerium** isotope (Ce-144) shows a difference of -70.9%. The relatively large difference and the fact that the difference is negative again indicates that the calculation underpredicts the measured concentration. The Ce-144 isotope is radioactive with a short half-life (284.6 days). If indeed, the measurement value is accurate, potential problems in the calculation could be the result of poor fission product yields or a radiative capture cross section that is too large, or both. In any event, the negative difference is consistent with that obtained in Rod N calculations.

The **zirconium** isotope (Zr-95) shows almost a 100% difference and in this case, the calculated value is almost negligible compared to the measured mass. The large difference between the calculated and measured values is explained in some detail in section 5.1.

Table 11 shows a comparison of the measured and calculated uranium isotopics by percentage of the total uranium mass. The percentages are in good agreement, particularly for the U-233, U-

234, U-235, and U-236 uranium isotopes with differences of only -0.1, +0.5, -1.2, and +0.5%, respectively. U-232 and U-238 are in reasonable agreement with differences of +35.3 and +47.3%, respectively. Again, these two uranium isotopes (U-232 and U-238) have positive differences and therefore represent conservative estimates.

Table 12 shows the effect on the calculated mass isotopics as a function of the number of cross section updates over the LWBR operation cycle. The total integrated burnup for each case is fixed at 7,000 MWd/MTHM. Of particular interest is the small improvement or convergence in the uranium isotopic concentrations as the number of cross section updates increases. Similarly, note the relatively minor changes in the fission product concentrations as a function of cross section updates. This is indicative of relatively constant neutron cross sections as a function of burnup, plus the fact that the relatively low burnup in this segment would also signal a weak dependency on the cross sections.

Again, our “best” estimate is obtained with the maximum number of cross section updates (4), but notice how well the single update or BOL cross sections predict the isotopic masses relative to the higher number of update cases. In addition, relative to the 4 update case, the differences between the calculated and measured uranium isotopic masses for the single update are all positive or conservatively estimated. On the other hand, the fission products are all under predicted with the exception of Xe-130 and Xe-131. The krypton isotopes, although, exhibit negative differences, they are all less than 4%, with the exception of Kr-82. This observation provides us with a reasonable justification for the defense of the calculated LWBR module radionuclide inventories based on BOL cross sections only. In order to ensure conservatism in these LWBR radionuclide calculations, small multipliers (e.g. 1.2 - 2.0) applied to the overall inventory (particularly the fission products) might suffice.

5.3 Rod H Segment 7 Results Comparison

For the reflector blanket fuel rod (Rod H), the calculated EOL uranium and fission product isotopic mass concentrations are compared with the measured assay EOL isotopics in Tables 13 and 14. The tables also list the BOL Th-232 loading for segment 7 for comparison. Note: the thorium reflector rods contained no uranium at BOL.

Table 13 presents a comparison of the BOL, measured EOL assay, and calculated EOL uranium, krypton, xenon, cesium-137, cerium-144, and zirconium-95 isotopic masses for segment 7 of Rod H. The far column labelled “DIFF” is the difference between the calculated and measured values

Table 12. Comparison of calculated uranium and fission product mass concentrations as a function of the number of cross section updates (Rod I).
(Burnup is fixed at 7,000 MWd/MTHM)

ISOTOPE	BOL (g)	ASSAY EOL (g)	1 update		2 updates		4 updates	
			CALC EOL (g)	DIFF* EOL (%)	CALC EOL (g)	DIFF* EOL (%)	CALC EOL (g)	DIFF* EOL (%)
U-232	0.000000	0.003327	0.004849	45.8	0.004904	47.4	0.004842	45.6
U-233	9.550038	8.735400	9.084000	4.0	9.093000	4.1	9.109000	4.3
U-234	0.125418	0.476050	0.491000	3.1	0.495000	4.0	0.499400	4.9
U-235	0.008750	0.051923	0.059210	14.0	0.056000	7.9	0.053580	3.2
U-236	0.001945	0.003438	0.003699	7.6	0.003649	6.1	0.003610	5.0
U-238	0.035972	0.021687	0.033290	53.5	0.033290	53.5	0.033310	53.6
Kr-82	0.000000	0.000137	0.000073	-46.8	0.000073	-46.4	0.000073	-46.7
Kr-83	0.000000	0.010654	0.010980	3.1	0.011000	3.2	0.011010	3.3
Kr-84	0.000000	0.021145	0.020750	-1.9	0.020740	-1.9	0.020710	-2.1
Kr-85	0.000000	0.004109	0.003961	-3.6	0.003961	-3.6	0.003959	-3.7
Kr-86	0.000000	0.034838	0.034550	-0.8	0.034550	-0.8	0.034530	-0.9
Xe-128	0.000000	0.000447	0.000351	-21.4	0.000354	-20.9	0.000338	-24.5
Xe-130	0.000000	0.000454	0.000680	49.7	0.000682	50.2	0.000683	50.3
Xe-131	0.000000	0.052169	0.051390	-1.5	0.052400	0.4	0.052870	1.3
Xe-132	0.000000	0.104670	0.092570	-11.6	0.091560	-12.5	0.091110	-13.0
Xe-134	0.000000	0.120770	0.108400	-10.2	0.108400	-10.2	0.108400	-10.2
Xe-136	0.000000	0.190040	0.145000	-23.7	0.144700	-23.9	0.144300	-24.1
Cs-137	0.000000	0.109590	0.108200	-1.3	0.108200	-1.3	0.108200	-1.3
Ce-144	0.000000	0.007790	0.002271	-70.8	0.002271	-70.8	0.002270	-70.9
Zr-95	0.000000	0.000042	2.952E-07	-99.3	2.952E-07	-99.3	2.953E-07	-99.3

* End-of-Life Difference = DIFF = ((Calc-assay)/Assay) * 100

Table 13. Comparison of measured versus calculated uranium and fission product mass isotopics for the BOL cross section update only and 3,515 MWd/MTHM burnup (Rod H).

ISOTOPE	BOL (g)	ASSAY EOL (g)	Experimental Error (g)	CALC EOL (g)	DIFF* EOL (%)
Th-232	423.65307	NA	NA	418.6	NA
U-232	0.000000	0.004893	0.000152	0.002166	-55.7
U-233	0.000000	3.661613	0.000791	3.409000	-6.9
U-234	0.000000	0.197812	0.000050	0.190700	-3.6
U-235	0.000000	0.021134	0.000222	0.017490	-17.2
U-236	0.000000	0.000833	0.000004	0.000708	-15.0
U-238	0.000000	0.000185	0.000194	4.717E-08	-100.0
Kr-82	0.000000	NA	NA	0.000023	NA
Kr-83	0.000000	0.005822	0.000805	0.005105	-12.3
Kr-84	0.000000	0.011066	0.001529	0.010220	-7.6
Kr-85	0.000000	0.002331	0.000330	0.002030	-12.9
Kr-86	0.000000	0.019024	0.002623	0.016680	-12.3
Xe-128	0.000000	NA	NA	0.000081	NA
Xe-130	0.000000	NA	NA	0.000303	NA
Xe-131	0.000000	0.025965	0.003597	0.025790	-0.7
Xe-132	0.000000	0.043673	0.006054	0.043800	0.3
Xe-134	0.000000	0.054583	0.007561	0.052260	-4.3
Xe-136	0.000000	0.085115	0.011789	0.084950	-0.2
Cs-137	0.000000	0.053351	0.000183	0.053310	-0.1
Ce-144	0.000000	0.004201	0.000024	0.001981	-52.8
Zr-95	0.000000	0.000023	4.367E-07	5.903E-07	-97.5

* End-of-Life Difference = DIFF = ((Calc-Assay)/Assay) * 100

NA = measured assay values not available.

Table 14. Comparison of measured versus calculated uranium mass isotopic percentages for the BOL cross section update only and 3,515 MWd/MTHM burnup (Rod H).

ISOTOPE	BOL (%)	ASSAY EOL (%)	CALC EOL (%)	DIFF* EOL (%)
U-232	0.00	0.13	0.06	-52.5
U-233	0.00	94.21	94.17	0.0
U-234	0.00	5.09	5.27	3.5
U-235	0.00	0.54	0.48	-11.2
U-236	0.00	0.02	0.02	-8.8
U-238	0.00	4.76E-03	1.30E-06	-100.0

* End-of-Life Difference = DIFF = ((Calc-Assay)/Assay) * 100

and is defined as: $((\text{Calc}-\text{Assay})/\text{Assay}) \times 100$. With this definition, a negative difference value indicates that the calculation under predicted the measured assay value, and a positive difference value indicates that the calculation overpredicted the measured assay value. Also included in a column to the right of the "ASSAY EOL" (or assay measurement at EOL) are the measurement errors associated with the measured mass values. Note that only 1 cross section update was performed for the calculation here, namely BOL cross sections.

Again, because the burnup data were not provided along with measured isotopic mass values, the burnup assumed in the calculation was achieved when the measured mass value of Cs-137 was matched. In Table 13, the calculated Cs-137 mass value is very close to the measured Cs-137 mass value (i.e. a difference of only -0.1%). This resulted in a burnup estimate of approximately 3,515 MWd/MTHM for segment 7 of Rod H.

There appears to be good agreement between the measured and calculated **uranium** isotopic masses. The U-233 mass difference is only -6.9%. The U-234, U-235, U-236 concentrations are quite good with differences of -3.6, -17.2, -15.0% differences, respectively. The U-232 and U-238 however deviate significantly from the other uranium isotopes, with differences of -55.7 and 100.0%, respectively. It is of interest to note here that the experimental error associated with the U-238 measurement is more than 100%! Hence, one would have little confidence in the measured U-238 concentration. Note all of the calculated uranium isotopic concentrations underpredict the measured concentration values and are therefore not considered to be conservative.

The **krypton** isotopes (Kr-83, Kr-84, Kr-85, Kr-86) have differences of less -13%, which represents good agreement, but since the differences are all negative would indicate slight underprediction of the fission product concentrations. Measured data for Kr-82 was not available for this rod segment.

The **xenon** isotopes (Xe-131, Xe-132, Xe-134, Xe-136) show very good agreement with differences of less than -5%. Measured data for Xe-128 and Xe-130 not available for this rod segment.

The **cesium** isotope (Cs-137) mass value reported in Table 13 shows a difference of only -0.1%. This difference is very small because it was used to "match" the calculation to measurement, and hence one would expect a zero difference with successive iterations in the approach to an exact matching. The Cs-137 isotope was chosen to match the calculation and measurement values for the reasons given in section 1.0 and at the same time provided what the author believes is a very

reasonable estimate of the burnup. The measured value has a very small experimental error and confidence in this measured value is expected to be relatively high.

The **cerium** isotope (Ce-144) shows a difference of -52.8%. The relatively large difference and the fact that the difference is negative again indicates that the calculation underpredicts the measured concentration. The Ce-144 isotope is radioactive with a short half-life (284.6 days). If indeed, the measurement value is accurate, potential problems in the calculation could be the result of poor fission product yields or a radiative capture cross section that is too large, or both. In any event, the negative difference is consistent with that obtained for rods N and I.

The **zirconium** isotope (Zr-95) shows almost a 100% difference and in this case, the calculated value is almost negligible compared with the measured mass. The large difference between the calculated and measured values is explained in some detail in section 5.1 above.

Table 14 shows a comparison of the measured and calculated uranium isotopics by percentage of the total uranium segment mass. The percentages are in good agreement, particularly for the U-233 with a rounded-off zero percent difference. The U-234, U-235, and U-236 uranium isotopes have differences of +3.5, -11.2, and -8.8%, respectively. The U-232 and U-238 again exhibit differences of -52.5 and -100.0%, respectively.

5.4 Radionuclide Inventories

From the ORIGEN2 output, a complete list of actinides and fission product nuclide concentrations are available. A listing of 145 of the major radionuclides are given in Appendix A for Rod N segment 3, Rod I segment 2, and Rod H segment 7, respectively. Note: these inventories are only for the segments and not the complete fuel rod or module. In addition, the activation products from the urania and thoria impurities are not included. The radionuclide concentrations are given in terms of "grams/segment" on a single date that corresponds to the midpoint of the destructive examination periods (sections 4.2, 4.3, and 4.4).

5.5 Decay Heats

The individual segment decay heats that correspond to the rod segment radionuclide inventories of section 5.4 (Appendix A) are in Table 15.

Table 15. Fuel rod segment decay heats.

Rod/Segment	Decay Heat (watts/segment)
Rod N segment 3	0.45
Rod I segment 2	0.23
Rod H segment 7	0.14

These decay heats again correspond to the midpoint date of the destructive examination period.

6.0 CONCLUSIONS

The end-of-life uranium isotopic and fission product mass concentrations measured from destructive chemical assay techniques on selected LWBR fuel rods were compared to calculated values using the calculational methodology discussed in this report. Three different LWBR fuel rods from three different module types were calculated and compared.

Several important conclusions can be drawn from the comparisons between calculated and measured LWBR uranium and fission product mass concentrations in the three different LWBR fuel rods evaluated, namely, Rod N (seed), Rod I (standard blanket), and Rod H (reflector):

(1) Excellent agreement was obtained in all three rods for the U-233, U-234, and U-235 concentration mass estimates. These three primary **uranium isotopes** account for greater than 99.5% of the EOL uranium mass in all three fuel rods. The percent differences between the calculated and measured values of U-233, U-234, and U-235 are:

Rod N segment 3: +1.5, +4.5, and +12.7%;

Rod I segment 2: +4.3, +4.9, and +3.2%; and

Rod H segment 7, -6.9, -3.6, and -17.2%, respectively.

The differences are relatively small considering the assumptions made in the calculations. The other three uranium isotopes (U-232, U-236, U-238) which together account for less than 0.5% of the total EOL uranium mass in the segments were in reasonable agreement. Because of the low mass concentrations of these three uranium isotopes, particularly in the reflector rod, some difficulty in obtaining accurate measured values (relatively large experimental errors) leads to the relatively larger percent differences observed (e.g. U-238 in the Rod H).

(2) Very good agreement (<20% difference) was obtained in all three rods for the four primary **krypton fission product isotopes**: Kr-83, Kr-84, Kr-85, and Kr-86, which together account for greater than 99.8% of the total EOL krypton mass in all three fuel rod segments. The percent differences between the calculated and measured values for these four krypton isotopes are:

Rod N segment 3: +8.6, +21.0, +13.3, and +16.5%;

Rod I segment 2: +3.3, -2.1, -3.7, and -0.9%; and

Rod H segment 7: -12.3, -7.6, -12.9, and -12.3%, respectively.

The fifth krypton isotope (Kr-82) accounts for less than 0.2% of the total EOL krypton mass in the segments were in reasonable, but poorer agreement. This isotope is produced by a complex

production chain and possible reasons for the poorer agreement are discussed in some detail in the section 5.1.

(3) Good agreement (<30% difference) was obtained in all three rods for the four primary **xenon fission product isotopes**: Xe-131, Xe-132, Xe-134, and Xe-136, which together account for greater than 99.8% of the EOL xenon mass in all three fuel rods. The percent difference between the calculated and measured values for these four xenon isotopes are:

Rod N segment 3: +4.2, +29.3, +20.3, and +21.1%;

Rod I segment 2: +1.3, -13.0, -10.2, and -24.1%; and

Rod H segment 7: -0.7, +0.3, -4.3, and -0.2%, respectively.

The other two xenon isotopes (Xe-128 and Xe-130) account for less than 0.2% of the total EOL xenon mass in the segments and are also in reasonable, but poorer agreement. These two xenon isotopes are not direct-yield fission products but rather are produced via transmutation reactions with other nearby fission products. Therefore, because these two xenon isotopes are dependent on other fission product concentrations and are produced via complex production chains, the additional computational complexity required to calculate these isotopes typically results in poorer agreement with the measured values. Further discussion of these two isotopes can be found in section 5.1.

(4) The EOL measured **Cs-137 fission product** concentration was used to match the depletion calculation or normalize the segment powers over the LWBR operating cycle. Hence, theoretically the agreement between calculation and measured value would be exact. However, due to the iterative nature of the calculation and the small, but insignificant residual convergence error, there remains a slight difference in the reported values.

(5) The **Ce-144 fission product** was in reasonable agreement between calculation and measurement with differences ranging between -50 and -70%. The calculation seems to under-predict this fission product for all three LWBR rods.

(6) The calculated concentrations for the **fission product Zr-95** were not on a one-to-one basis for comparison to the measured Zr-95 mass concentrations, because the calculated Zr-95 takes into account only the Zr-95 produced in the oxide fuel region as a fission product, whereas, the measured Zr-95 includes not only the mass in the oxide fuel, but also the mass in the irradiated segment clad. During the dissolution process, both the oxide fuel and the clad were dissolved together. The measured Zr-95 mass concentration is therefore driven by the far greater amount of the Zr-95 resulting from the dissolution of the irradiated Zircaloy-4 clad. This is indicative of the

assay results. The calculations were complete before it was determined that the cladding was dissolved with the fuel. In any event, the Zr-95 mass is small, because the half-life is very short (64.02 days). The decay time, i.e. the time elapsed since the LWBR rods were removed from the core until the time of dissolution and measurement, amounts to a duration of approximately 15 half-lives or a $3.1\text{E-}5$ reduction in the concentration. Even with the Zr-95 from the clad included in the measured mass value, the concentration is very low (20-40 micrograms/segment).

(7) The three rod segment burnups, based on calculated segment power outputs and normalized to the measured EOL Cs-137 mass, are given in Table 16.

Table 16. Calculated burnup estimates for the fuel rod segments.

Module	Rod	Segment	Burnup (MWd/MTHM)
Seed	N	3	37,330
Standard Blanket	I	2	7,075
Reflector	H	7	3,515

(8) The overall excellent to reasonable agreement in the comparisons between calculated and measured segment isotopic concentrations leads one to conclude that the methodology and computer model input data (i.e. the simplified reactor physics models and assumptions) were sufficient and can be relied upon to produce reasonably accurate radionuclide inventories for the complex LWBR core. The neutron cross sections developed for the LWBR using the methodology described herein apparently capture the important neutron spectral and spatial information in the LWBR core.

(9) Greater calculational accuracy or better agreement with measured data is obtained for some radionuclides by increasing the number of **cross section updates** over the LWBR operating lifetime. Additional updates tend to incorporate core physics changes that effect the local neutron flux and energy spectrum and hence the depletion calculation accuracy. The improvement, while small, is still perceptible for the LWBR fuel rods evaluated here. However, because it is relatively small, particularly when one compares, for example, Rod N calculated masses with 8 updates versus only one update (i.e. use of just the BOL cross sections), this lends credence to radionuclide inventories generated solely with BOL neutron cross sections.

(10) Typically, the more **complex the production chain(s)** to produce a certain isotope, the greater the percent difference between calculation and measurement. In particular, this applied to the Kr-82, Xe-128, and Xe-130 isotopes, but in general this conclusion also applies to both the uranium isotopes and the fission product isotopes, as well. It should be noted too that the more complex the production pathway for a particular isotope, the lower the EOL mass concentration is for that isotope.

(9) As a final conclusion, the isotopic mass percent differences in this report for the LWBR are comparable in magnitude to differences encountered with a previous validation effort reported in Reference 1 for commercial Light Water Reactor (LWR) spent nuclear fuels. In the LWR study, the same methodology as used herein was employed and compared with not only chemical assay data, but also to another and comparable calculational methodology involving the SCALE suite of codes.

9.0 REFERENCES

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APPENDIX A-1

Radionuclide Inventory for Rod N Segment 3 145 Isotope List*

*** Does not include activation products from impurities in the oxide fuel.**

**Table A-1. Isotopic Mass Concentrations in GRAMS
for the LWBR Rod N Segment 03.**

Information:

Units: GRAMS

Burnup: 37,000 MWd/MTHM
 BOL U-233: 8.9599 grams U-233 per element
 BOL U-234: 0.1177 grams U-234 per element
 BOL U-235: 0.008209 grams U-235 per element
 BOL U-236: 0.001824 grams U-236 per element
 BOL U-238: 0.033767 grams U-238 per element
 BOL Th-232: 142.3946 grams Th-232 per element
 Segment Volume: 17.3035 cm³
 Fuel Meat: UO₂-ThO₂ (Urania-Thoria)
 Fuel Meat: High Zone - Seed Module
 Fuel Enrichment: 98.23 wt% U-233
 Clad: Zircaloy-4

DECAY DATE: July 16, 1985

ISOTOPE	grams/segment
H 3	7.574E-06
BE 10	2.247E-08
C 14	4.542E-09
CL 36	0.000E+00
CR 51	0.000E+00
MN 54	0.000E+00
FE 55	0.000E+00
FE 59	0.000E+00
CO 60	0.000E+00
NI 59	0.000E+00
NI 63	0.000E+00
ZN 65	0.000E+00
SE 79	2.855E-03
KR 85	7.225E-03
RB 87	8.852E-02
SR 89	4.500E-09
SR 90	1.322E-01
Y 90	3.315E-05
Y 91	3.805E-08
ZR 93	1.610E-01
ZR 95	1.165E-07
NB 93M	3.306E-07
NB 94	5.357E-07
NB 95	1.420E-07
NB 95M	4.873E-11
MO 93	0.000E+00
TC 99	1.058E-01
RU103	2.128E-11
RU106	3.184E-04
RH103M	1.903E-14
RH106	2.993E-10
PD107	3.761E-03
AG110	1.457E-14
AG110M	9.613E-07

ISOTOPE	grams/segment
AG111	8.717E-47
CD113M	1.064E-05
CD113	1.542E-05
CD115M	2.495E-13
IN114	9.258E-20
IN114M	5.756E-15
IN115M	7.046E-20
SN119M	1.161E-07
SN121M	1.214E-06
SN123	1.272E-07
SN125	2.192E-37
SN126	7.919E-03
SB124	6.134E-11
SB125	1.139E-03
SB126	3.761E-10
SB126M	2.860E-12
TE123M	4.299E-10
TE125M	1.594E-05
TE127	1.451E-09
TE127M	4.145E-07
TE129	1.747E-16
TE129M	1.866E-13
I129	4.997E-02
I131	1.130E-41
XE131M	6.885E-31
XE133	7.243E-61
CS134	6.942E-03
CS135	1.461E-01
CS136	6.340E-28
CS137	2.009E-01
BA136M	2.843E-35
BA137M	3.074E-08
BA140	2.512E-27
LA140	3.788E-28
CE141	2.147E-12
CE142	2.320E-01
CE144	2.982E-03
PR143	6.249E-26
PR144	1.259E-07
PR144M	6.295E-10
ND144	1.925E-01
ND147	1.288E-31
PM145	0.000E+00
PM147	7.383E-03
PM148M	1.210E-11
PM148	8.863E-14
SM145	0.000E+00
SM147	1.996E-02
SM151	3.380E-03
EU152	6.614E-06
EU154	2.890E-03
EU155	9.382E-04
EU156	4.740E-25
GD153	4.786E-07
TB160	7.471E-11
TL206	3.352E-24
TL207	2.826E-13
TL208	2.246E-10
PB210	1.090E-09

ISOTOPE	grams/segment
PB211	2.186E-12
PB212	1.324E-07
BI211	1.290E-13
BI212	1.256E-08
PO212	6.647E-19
PO215	1.831E-18
PO216	5.285E-13
RN219	4.149E-15
RN220	1.995E-10
FR223	1.972E-14
RA223	1.054E-09
RA224	1.155E-06
RA226	7.503E-09
RA228	3.540E-08
AC227	7.640E-07
TH227	1.747E-09
TH228	2.235E-04
TH229	1.780E-04
TH230	1.701E-04
TH231	6.020E-13
TH232	1.390E+02
TH234	3.893E-13
PA231	8.432E-03
PA233	4.288E-11
PA234M	1.313E-17
PA234	5.863E-18
U232	1.092E-02
U233	5.747E+00
U234	7.643E-01
U235	1.480E-01
U236	1.336E-02
U237	1.701E-13
U238	2.681E-02
NP237	1.253E-03
PU236	4.414E-10
PU237	7.706E-17
PU238	2.862E-04
PU239	2.642E-03
PU240	4.879E-04
PU241	5.492E-04
PU242	1.011E-04
PU244	4.009E-09
AM241	1.109E-04
AM242M	1.470E-06
AM242	1.758E-11
AM243	1.708E-05
CM242	7.097E-08
CM243	1.608E-07
CM244	7.952E-06
CM245	6.941E-07
CM246	2.751E-08
CM247	3.921E-10

APPENDIX A-2

Radionuclide Inventory for Rod I Segment 2 145 Isotope List*

* Does not include activation products from impurities in the oxide fuel.

**Table A-2. Isotopic Mass Concentrations in GRAMS
for the LWBR Rod I Segment 02.**

Information:

Units: GRAMS

Burnup: 7,000 MWd/MTHM
 BOL U-233: 9.550038 grams U-233 per element
 BOL U-234: 0.125418 grams U-234 per element
 BOL U-235: 0.008750 grams U-235 per element
 BOL U-236: 0.001945 grams U-236 per element
 BOL U-238: 0.035972 grams U-238 per element
 BOL Th-232: 409.261910 grams Th-232 per element
 Segment Volume: 47.95 cm³
 Fuel Meat: UO₂-ThO₂ (Urania-Thoria)
 Fuel Meat: High Zone - Standard Blanket
 Fuel Enrichment: 98.23 wt% U-233
 Clad: Zircaloy-4

DECAY DATE: February 23, 1985

ISOTOPE	grams/segment
H 3	4.144E-06
BE 10	1.198E-08
C 14	2.421E-09
CL 36	0.000E+00
CR 51	0.000E+00
MN 54	0.000E+00
FE 55	0.000E+00
FE 59	0.000E+00
CO 60	0.000E+00
NI 59	0.000E+00
NI 63	0.000E+00
ZN 65	0.000E+00
SE 79	1.525E-03
KR 85	3.959E-03
RB 87	4.763E-02
SR 89	1.744E-08
SR 90	7.140E-02
Y 90	1.791E-05
Y 91	1.119E-07
ZR 93	8.706E-02
ZR 95	2.953E-07
NB 93M	1.670E-07
NB 94	3.005E-07
NB 95	3.599E-07
NB 95M	1.235E-10
MO 93	0.000E+00
TC 99	6.115E-02
RU103	1.407E-10
RU106	2.189E-04
RH103M	1.258E-13
RH106	2.057E-10
PD107	2.034E-03
AG110	4.095E-15
AG110M	2.702E-07

ISOTOPE	grams/segment
AG111	3.021E-41
CD113M	4.539E-06
CD113	2.023E-05
CD115M	1.255E-12
IN114	1.000E-19
IN114M	6.218E-15
IN115M	3.545E-19
SN119M	7.592E-08
SN121M	6.453E-07
SN123	1.466E-07
SN125	3.653E-33
SN126	4.205E-03
SB124	5.421E-11
SB125	6.718E-04
SB126	1.997E-10
SB126M	1.519E-12
TE123M	5.039E-11
TE125M	9.396E-06
TE127	1.945E-09
TE127M	5.555E-07
TE129	1.824E-15
TE129M	1.948E-12
I129	2.771E-02
I131	1.482E-36
XE131M	1.616E-27
XE133	7.023E-53
CS134	1.640E-03
CS135	9.009E-02
CS136	3.432E-25
CS137	1.082E-01
BA136M	1.539E-32
BA137M	1.655E-08
BA140	3.302E-24
LA140	4.980E-25
CE141	2.474E-11
CE142	1.236E-01
CE144	2.270E-03
PR143	5.255E-23
PR144	9.585E-08
PR144M	4.791E-10
ND144	9.450E-02
ND147	5.687E-28
PM145	0.000E+00
PM147	7.332E-03
PM148M	4.915E-11
PM148	3.600E-13
SM145	0.000E+00
SM147	1.799E-02
SM151	2.726E-03
EU152	1.343E-05
EU154	4.842E-04
EU155	2.021E-04
EU156	3.883E-23
GD153	2.595E-07
TB160	4.852E-11
TL206	1.174E-24
TL207	4.221E-13
TL208	9.700E-11
PB210	1.421E-10

ISOTOPE	grams/segment
PB211	3.265E-12
PB212	5.721E-08
BI211	1.927E-13
BI212	5.427E-09
PO212	2.871E-19
PO215	2.734E-18
PO216	2.283E-13
RN219	6.196E-15
RN220	8.619E-11
FR223	2.943E-14
RA223	1.574E-09
RA224	4.991E-07
RA226	6.130E-09
RA228	1.018E-07
AC227	1.140E-06
TH227	2.609E-09
TH228	9.653E-05
TH229	2.601E-04
TH230	1.493E-04
TH231	2.179E-13
TH232	4.062E+02
TH234	4.836E-13
PA231	1.235E-02
PA233	1.886E-11
PA234M	1.631E-17
PA234	7.283E-18
U232	4.842E-03
U233	9.109E+00
U234	4.994E-01
U235	5.358E-02
U236	3.610E-03
U237	3.219E-14
U238	3.331E-02
NP237	2.311E-04
PU236	3.566E-11
PU237	2.021E-17
PU238	2.729E-05
PU239	1.669E-03
PU240	2.145E-04
PU241	1.040E-04
PU242	7.820E-06
PU244	3.909E-11
AM241	2.005E-05
AM242M	1.545E-07
AM242	1.849E-12
AM243	5.896E-07
CM242	1.002E-08
CM243	4.049E-09
CM244	8.983E-08
CM245	2.678E-09
CM246	4.220E-11
CM247	1.915E-13

APPENDIX A-3

Radionuclide Inventory for Rod H Segment 7 145 Isotope List*

* Does not include activation products from impurities in the oxide fuel.

**Table A-3. Isotopic Mass Concentrations in GRAMS
for the LWBR Rod H Segment 07.**

Information:

Units: GRAMS

Burnup: 3,700 MWd/MTHM
 BOL U-233: 0.000000 grams U-233 per element
 BOL U-234: 0.000000 grams U-234 per element
 BOL U-235: 0.000000 grams U-235 per element
 BOL U-236: 0.000000 grams U-236 per element
 BOL U-238: 0.000000 grams U-238 per element
 BOL Th-232: 423.650510 grams Th-232 per element
 Segment Volume: 49.41 cm³
 Fuel Meat: ThO₂ (Thoria only)
 Fuel Meat: Thoria Reflector Blanket
 Fuel Enrichment: Natural Thorium
 Clad: Zircaloy-4

DECAY DATE: January 31, 1985

ISOTOPE	grams/segment
H 3	2.108E-06
BE 10	5.770E-09
C 14	1.166E-09
CL 36	0.000E+00
CR 51	0.000E+00
MN 54	0.000E+00
FE 55	0.000E+00
FE 59	0.000E+00
CO 60	0.000E+00
NI 59	0.000E+00
NI 63	0.000E+00
ZN 65	0.000E+00
SE 79	7.344E-04
KR 85	2.030E-03
RB 87	2.298E-02
SR 89	4.453E-08
SR 90	3.523E-02
Y 90	8.836E-06
Y 91	2.433E-07
ZR 93	4.203E-02
ZR 95	5.903E-07
NB 93M	6.662E-08
NB 94	1.461E-07
NB 95	7.193E-07
NB 95M	2.470E-10
MO 93	0.000E+00
TC 99	2.964E-02
RU103	5.089E-10
RU106	1.758E-04
RH103M	4.551E-13
RH106	1.652E-10
PD107	9.804E-04
AG110	1.641E-15
AG110M	1.083E-07

ISOTOPE	grams/segment
AG111	2.154E-38
CD113M	2.167E-06
CD113	6.301E-06
CD115M	3.582E-12
IN114	8.239E-20
IN114M	5.122E-15
IN115M	1.012E-18
SN119M	6.503E-08
SN121M	3.144E-07
SN123	1.800E-07
SN125	8.897E-31
SN126	2.022E-03
SB124	5.451E-11
SB125	4.069E-04
SB126	9.605E-11
SB126M	7.305E-13
TE123M	1.580E-11
TE125M	5.691E-06
TE127	2.643E-09
TE127M	7.549E-07
TE129	8.508E-15
TE129M	9.086E-12
I129	1.335E-02
I131	8.007E-34
XE131M	1.584E-25
XE133	1.750E-49
CS134	8.694E-04
CS135	2.826E-02
CS136	1.603E-23
CS137	5.331E-02
BA136M	7.188E-31
BA137M	8.156E-09
BA140	2.319E-22
LA140	3.497E-23
CE141	1.209E-10
CE142	5.958E-02
CE144	1.981E-03
PR143	2.901E-21
PR144	8.367E-08
PR144M	4.182E-10
ND144	4.803E-02
ND147	7.347E-26
PM145	0.000E+00
PM147	4.717E-03
PM148M	6.607E-11
PM148	4.845E-13
SM145	0.000E+00
SM147	8.083E-03
SM151	5.307E-04
EU152	3.858E-06
EU154	2.482E-04
EU155	9.478E-05
EU156	1.955E-21
GD153	1.852E-08
TB160	3.913E-11
TL206	1.135E-25
TL207	2.439E-13
TL208	4.229E-11
PB210	5.609E-11

ISOTOPE	grams/segment
PB211	1.887E-12
PB212	2.494E-08
BI211	1.113E-13
BI212	2.366E-09
PO212	1.252E-19
PO215	1.580E-18
PO216	9.953E-14
RN219	3.580E-15
RN220	3.758E-11
FR223	1.701E-14
RA223	9.095E-10
RA224	2.176E-07
RA226	3.334E-09
RA228	1.047E-07
AC227	6.593E-07
TH227	1.507E-09
TH228	4.208E-05
TH229	7.055E-05
TH230	7.996E-05
TH231	7.110E-14
TH232	4.186E+02
TH234	2.119E-16
PA231	7.345E-03
PA233	1.542E-10
PA234M	7.145E-21
PA234	3.192E-21
U232	2.166E-03
U233	3.409E+00
U234	1.907E-01
U235	1.749E-02
U236	7.079E-04
U237	4.496E-19
U238	4.717E-08
NP237	1.561E-05
PU236	6.546E-13
PU237	1.050E-18
PU238	1.184E-06
PU239	8.052E-08
PU240	5.863E-09
PU241	1.452E-09
PU242	9.774E-11
PU244	3.150E-16
AM241	1.992E-10
AM242M	2.310E-13
AM242	2.764E-18
AM243	4.083E-12
CM242	9.352E-14
CM243	1.453E-14
CM244	3.609E-13
CM245	4.757E-15
CM246	1.517E-16
CM247	5.061E-19