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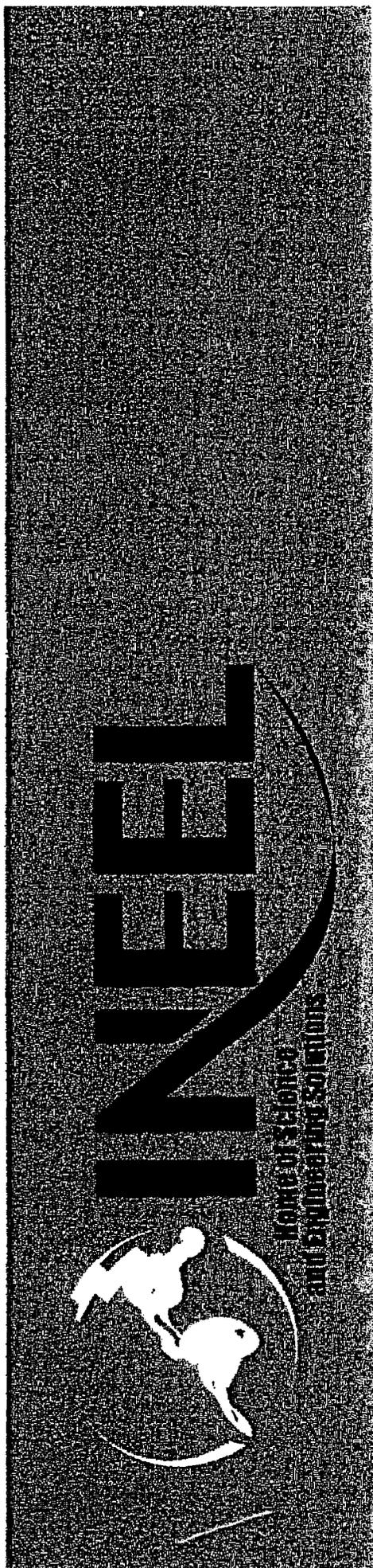
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Methodology to Determine Radioisotope Contents in RH TRU Waste Drums from Irradiated Fuel Examination at ANL-E

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September 2003



*Idaho National Engineering and Environmental Laboratory
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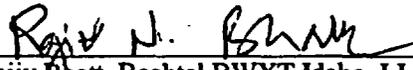
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ABSTRACT

This document outlines the methods to estimate radionuclide contents in remote-handled (RH)-transuranic (TRU) waste drums stored at the Idaho National Engineering and Environmental Laboratory. The methods are based on minimal obtainable acceptable knowledge information on the RH TRU waste generation process, the identities of the fuel elements destructively examined at the Argonne National Laboratory-East, and the design and irradiation history of those fuel elements. Because the RH TRU waste stored at the Idaho National Engineering and Environmental Laboratory constitutes a small fraction of the allowable RH TRU waste to be disposed at the Waste Isolation Pilot Plant, large uncertainties, up to a few hundred percent, in the estimates of the radionuclide contents are not expected to have any significant impact on the waste disposal limits at Waste Isolation Pilot Plant. The methods presented here are based on general physical and engineering principles and are not tailored to specific situations. Consequently, the results for any particular waste drum will have large, but still tolerable, uncertainties.

The methods provide estimates of: (1) total activity, (2) activities of ten reportable radionuclides (Am-241, Pu-238, Pu-239, Pu-240, Pu-242, U-233, U-234, U-238, Sr-90, and Cs-137), (3) TRU activity, (4) activities of the highest-hazard radionuclides that comprise more than 95% of the total radioactive hazard, and (5) the activity concentrations (Ci per unit mass of waste). These estimates are made at both the batch level and the individual drum level. Methods to calculate the uncertainties of the estimated parameters are also given.

As an example of the application of the methodology, radionuclide parameters for a batch of RH TRU waste drums numbered 728 through 737 are given in the Appendix.

SUMMARY

In 2001, the Department of Energy Carlsbad Field Office (DOE-CBFO) requested the Idaho National Engineering and Environmental Laboratory (INEEL) to develop a site-specific approach to the characterization of stored remote-handled transuranic (RH TRU) waste to support a Change Notice to the U.S. Environmental Protection Agency (EPA) for the 40 CFR 194 Certification of the Waste Isolation Pilot Plant (WIPP).^a This document is a response to that request.

The general RH TRU waste characterization program proposed by DOE-CBFO^b will rely on Acceptable Knowledge (AK) to obtain most of the needed characterization parameters. If the AK information was generated outside an American Society of Mechanical Engineers Nuclear Quality Assurance (NQA)-1 quality assurance program,^c the RH TRU waste characterization program will use the quality assurance provisions of 40 CFR 194.22(b) to qualify the AK information by one or more of the following methods:

- Peer review in accordance with Nuclear Regulatory Guide (NUREG)-1297^d
- Corroborating data
- Confirmatory testing
- Demonstrating the equivalence of an alternate quality assurance (QA) program to NQA-1.

The INEEL-specific RH TRU Radioactive Waste Characterization Program plans to follow the guidance of the general DOE-CBFO characterization program using AK as the primary means to characterize the RH TRU waste. Because the AK on the RH TRU waste stored at the INEEL was not generated under an NQA-1 quality assurance program or its equivalent, the INEEL plans to use the peer review option to qualify the AK information. In addition, certain characterization parameters will be generated based on AK information using proposed methods presented in this document. INEEL plans to have these methods peer reviewed and approved by the DOE-CBFO for the characterization of the RH TRU waste.

The Waste Stream

The INEEL has in storage approximately 700 containers of RH TRU waste to be disposed of at WIPP. Of these 700 containers, 617 are 30-gal drums containing RH TRU waste shipped from the Argonne National Laboratory-East (ANL-E) to the INEEL in 23 batches. The waste was generated from the destructive examination of experimental fuel elements (cylindrical pins) irradiated in the Experimental Breeder Reactor II (EBR-II) reactor at ANL-West (ANL-W) from the 1970s to the mid-1990s. The irradiated experimental fuel elements contained uranium, plutonium, other transuranic elements, activation products, and fission products. The destructive examination of the fuel elements involved

a. Inez R. Triay letter to Lori Fritz, "Request for Site Support to the Proposed Changes to the U. S. Environmental Protection Agency's Waste Isolation Pilot Plant 40 CFR Part 194 Certification," July 5, 2001.

b. *Notification of Planned Change to the Environmental Protection Agency (EPA) 40 CFR PART 194 Certification of the Waste Isolation Pilot Plant*, DOE/WIPP-DRAFT-3213, Revision 2.

c. NQA-1, *Quality Assurance Program for Nuclear Facility Applications*, ASME Nuclear Standards.

d. *Peer Review for High-Level Nuclear Waste Repositories*, NUREG-1297, Nuclear Regulatory Commission, February 1988.

sectioning the fuel elements into samples and grinding and polishing the samples for examination. These operations generated a fine particulate, or swarf, of fuel and cladding material, most of which was cleaned up and stored in the hot cell at ANL-E. The residual contamination in the hot cell was periodically cleaned with rags and tissue paper, which was placed in the waste drums. Occasionally, contaminated tools, such as grinding and cutting wheels, were also packed into waste drums, along with contaminated glassware, light bulbs, and other noncombustible material. Based on the level of radioactive contents in the waste, the waste is classified as RH TRU waste. The physical and radioactive characteristics of the entire waste stream (617 30-gal drums) at the time of shipping from ANL-E to the INEEL is shown in the table below.

INEEL inventory of RH TRU waste shipped from ANL-E.

Total volume	70 m ³
Number of Containers	617 30-gal drums
Total activity	1,295 Ci
Total TRU activity	26 Ci
Total Pu + U-235	855 g
Average total activity per drum	2.10 Ci
Average TRU activity per drum	0.04 Ci or 900 nCi/g
Average Pu + U-235 per drum	1.39 g
Container surface dose rate range	200 mR/hr to 30 R/hr

The total allowed RH TRU waste volume to be disposed of at WIPP is approximately 4% of the total TRU (contact-handled plus remote-handled) waste volume (6.2 million ft³). The Land Withdrawal Act (LWA) limits the total radioactivity in the RH TRU waste emplaced in WIPP to 5.1 million Curies.^e This total allowed RH TRU activity is expected to be less than 10% of the total radioactivity emplaced in the repository at the time of its closure in 2033.^f The volume of the INEEL RH TRU waste from ANL-E is less than 1% of total WIPP-bound RH TRU waste volume and contained at the time of its shipping to the INEEL (before the mid-1990s) less than 0.03% of the LWA RH TRU activity limit. The waste also contains too little fissile material (Pu-239, Pu-241, and U-235) to cause criticality concerns.

Approach to Radiological Characterization

For the RH TRU waste generated at ANL-E and stored at INEEL, the dominant TRU contents in the waste drums are documented in official records. The basis for the TRU contents was radionuclide inventory information on fuel elements examined, hot cell procedures, and estimates of waste generation based on operational experience. Also, at the time these drums were shipped to the INEEL, dose rates were measured around the drums. From these dose rate measurements, the Cs-137 contents in the drums were estimated and documented on the waste shipment forms. These documented radionuclide contents provide important, but not all, radiological characterization parameters required under the proposed WIPP RH TRU waste acceptance criteria (WAC).^g However, the documented information and supplemental information on the history of the destructively examined fuel elements, here collectively referred to as the AK information, provide sufficient information for estimating the radionuclide contents in the waste drums as required by the WAC.

e. *The Waste Isolation Pilot Plant Land Withdrawal Act, as amended*, Public Law 102-579, September 23, 1996.

f. M. K. Knowles and P. E. Schoemaker, *RH TRU Inventory Impact Assessment Report*, April 30, 2001.

g. *Remote-Handled Waste Acceptance Criteria for the Waste Isolation Pilot Plant*, DOE/WIPP-02-3123, Rev.0, June 2002; effective date: TBD.

This document presents methods by which the radionuclide contents and other radiological parameters not included in the AK information are estimated. The methods rely on knowledge of material origins and basic physical principles, but specific information about the waste will be used whenever it is obtainable. When general knowledge and basic physical principles are used instead of specific information, estimated parameters will have wider bands of uncertainty than when specific information is used. However, such added uncertainties in estimated parameters are small enough that they will have no impact on meeting the requirements of the WAC.

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ACRONYMS

AGHCF	Alpha-Gamma Hot Cell Facility
AK	acceptable knowledge
ANL-E	Argonne National Laboratory-East
ANL-W	Argonne National Laboratory-West
ASME	American Society of Mechanical Engineers
BU	burnup
CBFO	Carlsbad Field Office
DOE	Department of Energy
DOE-CBAO	Department of Energy-Carlsbad Area Office
EBR-II	Experimental Breeder Reactor II
EPA	Environmental Protection Agency
INEEL	Idaho National Engineering and Environmental Laboratory
LWA	Land Withdrawal Act
NQA	Nuclear Quality Assurance
NUREG	Nuclear Regulatory Guide
QA	Quality Assurance
RH TRU	remote-handled transuranic
RWMC	Radioactive Waste Management Complex
TRU	transuranic
WAC	waste acceptance criteria
WIPP	Waste Isolation Pilot Plant

Methodology to Determine Radioisotope Contents in RH TRU Waste Drums from Irradiated Fuel Examination at ANL-E

1. PURPOSE

Most of the remote-handled transuranic (RH TRU) waste generated in the Alpha-Gamma Hot Cell Facility (AGHCF) at the Argonne National Laboratory-East (ANL-E) in Chicago from 1976 to 1995 is being stored at the Radioactive Waste Management Complex (RWMC) at the Idaho National Engineering and Environmental Laboratory (INEEL). The origin of most of the waste can be traced to fuel elements that underwent irradiation in the Experimental Breeder Reactor II (EBR-II) reactor at the Argonne National Laboratory-West (ANL-W) in Idaho. These fuel elements were fabricated from materials that came from defense-related programs and so the waste that was generated from their destructive examination in the AGHCF, based on the WIPP Land Withdrawal Act¹ (LWA) of 1992, as amended, is eligible for disposal at the Waste Isolation Pilot Plant (WIPP).

Before these RH TRU waste drums stored at the RWMC can be disposed of at the WIPP, a number of conditions must be met to satisfy the waste acceptance criteria (WAC) set by the U.S. Department of Energy Carlsbad Field Office (DOE-CBFO).² These acceptance criteria include those for the payload container and the physical, chemical, and radiological properties of the waste. Under these criteria, a set of requirements must be met. These requirements include operations and safety requirements, transportation safety requirements, hazardous waste facility permit requirements, compliance certification decision requirements, and LWA requirements. These criteria and requirements are traceable to laws and regulations such as the LWA and the U.S. Environmental Protection Agency (EPA) regulations 40 CFR 191 and 40 CFR 194.

The AGHCF waste shipped to and stored at the INEEL was collected and packaged before the implementation of quality assurance programs that would have complied with the quality assurance requirements in 40 CFR 194.22. However, evaluations of Acceptable Knowledge (AK) information show that enough data and information exist to allow estimates of radionuclide contents in the drums to satisfy the characterization requirements without recourse to sampling the drum contents for radiochemical assay. The purpose of this paper is to define a methodology to achieve such estimates based on AK and other supplemental information to meet the DOE RH TRU WAC in terms of radiological properties of the waste. Specifically, the required radionuclide characterizations are:

1. Total activity
2. Activities of 10 specific radionuclides: Am-241, Pu-238, Pu-239, Pu-240, Pu-242, U-233, U-234, U-238, Sr-90 and Cs-137
3. TRU activity, defined as activities of alpha-emitting transuranic radionuclides with half-lives longer than 20 years
4. Activities of the highest-hazard radionuclides that combine to give at least 95% of the total radioactive hazard
5. Activity concentration
6. Surface dose rate of waste containers.

2. RADIOISOTOPES TO BE CONSIDERED

Based on the characterization of radionuclides in a batch of RH TRU waste drums that originated from ANL-E and were stored at the INEEL,³ approximately 13 years after the end of irradiating the fuel elements from which the waste was generated, over 95% of the total activity of the waste came from only a few radioisotopes. These isotopes are Cs-137, its decay daughter Ba-137M, Sr-90, its decay daughter Y-90, Pu-241, and the transuranic isotopes Pu-239, Pu-240, and Am-241 (hereafter referred to as "significant isotopes" or "significant radionuclides"). With the passage of time, but before most of the Cs-137 and Sr-90 will have decayed in a few hundred years, (i.e., within the time frame that the waste will be disposed of at the WIPP) the proportion of activity from these significant isotopes will increase further because of the more rapid decay of the other isotopes (mostly fission and activation products) that comprise most of the remaining activity. It is therefore only necessary to consider these significant isotopes for determining the total activity in the waste.

In addition to determining the activities of the significant isotopes (Cs-137, Ba-137M, Sr-90, Y-90, Pu-241, Pu-239, Pu-240, and Am-241), and, hence, the total activity in the waste, the activities of a few other radionuclides are also required to be determined even though these additional radionuclides contribute only an insignificant fraction of the total activity in the waste. These additional radionuclides are U-233, U-234, U-238, Pu-238, and Pu-242 in the required 10-radionuclide list. Another radionuclide, Np-237, which is important to long-term hazard evaluation and is an intermediate product in the generation of Pu-238, will also be considered in this document.

3. ACCEPTABLE KNOWLEDGE AND OTHER SUPPLEMENTAL INFORMATION

The contents of RH TRU waste drums from the ANL-E were documented in waste shipment forms, such as the Waste Package Data Sheet and the Solid Radioactive Waste Disposal Requisition forms.⁴ These forms list the dominant TRU (Pu-239, Pu-240, and Pu-241) and Cs-137 contents of the waste drums. These forms, or records, constitute the core AK information. Other obtainable supplemental AK information includes the origin of the waste and the methods by which the radiological contents of the waste drums were estimated. The methods of calculation in this document will show that such information is sufficient for estimating the quantities of the other radionuclides not documented by ANL-E but required by the WAC. Specifically, such AK information includes:

1. **Origin of waste** – Records documenting the packaging of waste in drums for waste generated from the destructive examination of fuel elements in the hot cell. The records contain the beginning and end dates of waste generation, the identities of the fuel elements destructively examined in the hot cell during that time period, and identities of the waste drums.
2. **Fuel element design** – Fuel element design information includes the mass and chemical composition of the cladding and fuel, and the isotopic composition of uranium (U-235 and U-238) and plutonium (Pu-239 and Pu-240). The design information comes from a variety of sources – experimental design requirements, certification of as-built fuel elements, safety analysis reports, safeguard materials database, and other pertinent documents.
3. **Fuel element irradiation history** – Records of the burnup of the heavy metal (uranium and plutonium) contents of the fuel, or records containing sufficient information for the calculation of the burnup, and records showing the date the fuel element was last removed from the reactor.
4. **Fuel element destructive examination history** – Records showing the destructive examination dates of the fuel element in the hot cell.
5. **Waste packaging** – Mass and type (combustible or noncombustible) of waste in waste drums.

4. ESTIMATING RADIONUCLIDE ACTIVITIES

The methods discussed here to estimate the activities of radionuclides depend on knowledge of only a minimum set of parameters. These are the pre- and post-irradiation fuel composition that includes the masses of U-235, U-238, Pu-239, and Pu-240, irradiation time, and time at which the fuel was removed from the reactor. All such information is obtainable from the AK information. The masses give an estimate of the burnup of the fuel. The time parameters allow decay calculations for the short-lived radionuclides (half-life less than a few hundred years). Other parameters, such as detailed isotopic composition of the fresh fuel, if available, can be used to arrive at more accurate estimates of the small quantities of some special isotopes, but are not necessary for adequate overall characterization of the waste.

Summary descriptions of the methods used for estimating the various quantities are presented in this report. Detailed derivation of the methods and mathematical formulas can be found in two companion documents, *Evaluation of Radionuclide Contents in RH-TRU Waste Drums 728-737 Based on Reported Irradiated Fuel Examination*³ and "Quantifying Special Actinides in RH-TRU Waste from Irradiated Fuel Examination at ANL-E."⁵

4.1 Estimate of Burnup

Burnup (BU) is defined as fractional depletion of heavy metal mass (plutonium, uranium, and other actinides) in fuel due to fission of these isotopes. Because changes in the masses of actinides other than those of plutonium and uranium are small compared with those of plutonium and uranium, the burnup can be approximated as the ratio of the decrease in the mass of the plutonium and uranium in the fuel because of irradiation to the initial plutonium and uranium mass. The SPM-24 form⁴ gives the uranium and plutonium masses before and after irradiation. These values were derived from code calculations for the irradiation of the fuel element using the code package, BURNOUT,⁶ at ANL-W. A crucial parameter used to estimate the generation of the actinides due to fuel irradiation is the product of the mass-weighted fission cross-section and the neutron fluence.⁵ The relationship between this parameter and the burnup is defined in the following paragraph.

Let M_R denote the irradiated mass and M_0 denote the preirradiated mass. Then

$$M_R = M_0 \exp(-\sigma_f \Phi t) \quad (1)$$

where σ_f is the mass-weighted fission cross-section, Φ is the neutron flux, t is the irradiation time, and Φt is the neutron fluence. The burnup is given by

$$BU = (M_0 - M_R) / M_0 = 1 - \exp(-\sigma_f \Phi t) \quad (2)$$

The term, $\sigma_f \Phi t$, expressed as a function of the burnup, is

$$\sigma_f \Phi t = -\ln(1 - BU) = -\ln[1 - (M_0 - M_R) / M_0] \quad (3)$$

4.2 Activation Products Inventory

As shown in the *Evaluation* document³, the activities of the activation products 13 years after irradiation of the fuel elements comprise less than 0.12% of the total activities in the fuel elements. Almost all these activities come from Fe-55 (2.7 year half-life with no gamma ray in its decay) and Co-60

(5.3 year half-life with gamma rays at 1.17 and 1.33 MeV in its decay). The uncertainty in the Fe-55 generated in the cladding is only sensitive to the burnup of the fuel since the isotopic ratios of iron in the cladding can be accurately quantified. The Co-60 generated in the cladding, however, depends on the initial cobalt impurity in the cladding. The level of the cobalt impurity is small but its precise value is usually not obtainable from AK. For this reason, there can be large uncertainties in estimating the Co-60 inventory in the waste. If a waste drum is found to have excessive radiation levels beyond those calculated from AK for the Cs-137 contents, the drum will be gamma-scanned for the presence of Co-60. Otherwise, activation products will not be characterized for the RH TRU waste.

4.3 Fission Products Inventory

The fission product inventory in a fuel element approximately 13 or more years after irradiation is calculated from the burnup and the initial heavy metal mass of the fuel element. Over 95% of the fission product inventory can be accounted for by the activities of Cs-137, Sr-90, and their decay daughters in secular equilibrium with their respective parents (activities being equal for parent and daughter at any particular time).³

Regardless of the nuclei being fissioned and the energy of the neutrons causing the fission, the yield of Cs-137 is very close to 6.4 atomic percent per fission (the range being 6.2 to 6.6%). The fissionable nuclei in the EBR-II reactor include U-235, U-238, Pu-239, Pu-240, and Pu-241. The Cs-137 inventory will be calculated from the Cs-137 yield and the burnup of heavy metal, and decay-corrected to the time of characterization. The activity of Ba-137M is 94% of the Cs-137 activity.

The inventory of Sr-90 in a fuel element can be similarly estimated. However, the fission yield of Sr-90 depends on the nucleus being fissioned and, to a lesser extent, the neutron energy causing the fission. For example, the Sr-90 yield from U-235 fission by thermal neutrons is 5.8 atomic percent, while that from Pu-240 fission by neutrons having a fission energy spectrum (mostly high-energy neutrons around 1 MeV in energy) is only 1.8 atomic percent, so the Sr-90 yield is sensitive to the nuclei being fissioned. The average Sr-90 will be calculated as the fission-weighted average based on the initial masses of U-235, U-238, Pu-239, and Pu-240 and their fission rates. The inventory of Sr-90 will be calculated from the average fission yield and burnup of the fuel element, and decay-corrected to the time of characterization.

4.4 Actinides Inventory

An AK package for a batch of waste contains documents showing the quantities of U-235, U-238, Pu-239, and Pu-240 in the waste. These quantities will be attributed to the waste with no change, but their uncertainties will be calculated and reported. The quantities and their uncertainties of the other required heavy metal isotopes, Am-241, Pu-238, Pu-242, U-233, and U-234, together with Pu-241 and Np-237, (*special actinides* for short) will be calculated based on AK information. Detailed discussion on their calculations is contained in the companion document, "Quantifying Special Actinides in RH-TRU Waste from Irradiated Fuel Examination at ANL-E."⁵ The following subsections described the methods to quantify these special actinides based on results in that document.

For each batch of waste, the fuel elements associated with the generation of that waste are identified in its AK package. The AK also contains information from which the burnup of the fuel elements can be calculated (Equation 2). A conservative (higher than actual) estimate of the quantities of the special actinides in the fuel elements is to add the generated quantities of these actinides because of irradiation to the initial quantities of these actinides, but ignore any depletion of these actinides during irradiation. Quantities of the radionuclides are then only functions of their initial values, the generation and fission cross-sections, and the burnup. Uncertainties of these quantities can be estimated from these

functional relationships. The initial quantities and the generation paths are given below for each of the special actinides. Except for the initial values of U-235, U-238, Pu-239, and Pu-240, which will be the AK values, initial values used will be the greater of the estimated and AK values.

4.4.1 U-235, U-238, Pu-239, and Pu-240

As mentioned in the first paragraph of Section 4.4, quantities of these radionuclides will be obtained from AK documents.

4.4.2 U-233

U-233 was not present in the fresh experimental fuel irradiated in the EBR-II reactor. Three generation paths for U-233 will be considered. These are: (1) $(n,2n)$ reaction on U-234; (2) $(n,3n)$ reaction on U-235; and (3) two successive $(n,2n)$ reactions on U-235. The calculation of U-233 requires the initial values of U-235 (from AK) and U-234 (from either AK or estimate as given in Section 4.4.3).

4.4.3 U-234

The experimental EBR-II fuel all contained highly-enriched uranium. Because of the enrichment process, the ratio of U-234 to U-235 was enhanced over that in natural uranium. The initial mass of U-234 will be either 1% U-235⁷ or the value from available AK, whichever is greater. The generation path of U-234 to be considered is the $(n,2n)$ reaction on U-235.

4.4.4 Pu-238

Pu-238 was present in the fresh fuel that contained plutonium. The amount of initial Pu-238 is estimated from the Pu-240 to Pu-239 ratio or given by AK, whichever is greater.

The reaction paths to be considered in the generation of Pu-238 are: (1) $(n,2n)$ reaction on Pu-239; (2) activation of Am-241 to Am-242, which decays to Cm-242, then to Pu-238 in a short time; (3) three successive neutron captures by U-235; (4) two successive neutron captures by U-236; and (5) $(n,2n)$ reaction on U-238 followed by neutron capture. The initial values of U-235, U-238, and Pu-239 are given by AK. If the AK package gives a value for the U-236, that value will be used for the initial value of U-236; otherwise, the initial content of U-236 will be zero (no pre-irradiation of the uranium used to make the fresh fuel). The initial mass of Am-241 will be either given by AK or estimated as in Section 4.4.7.

4.4.5 Pu-241

Pu-241 was present in the fresh fuel when the fuel contained plutonium. The amount of initial Pu-241 will be either the estimated value from the Pu-240 to Pu-239 ratio, or given by AK, whichever is greater.

Pu-241 generation will be calculated from two successive neutron captures by Pu-239 and a neutron capture by Pu-240. The initial values of Pu-239 and Pu-240 are those contained in the AK package.

4.4.6 Pu-242

Pu-242 was present in the fresh fuel when the fuel contained plutonium. The generation of Pu-242 will be calculated from: (1) a neutron capture by Pu-241; (2) two successive neutron captures by Pu-240;

and (3) three successive neutron captures by Pu-239. The initial values of Pu-239 and Pu-240 will be given by AK and the initial value of Pu-241 is determined in Section 4.4.5.

4.4.7 Am-241

The initial mass of Am-241 in a fuel element will be calculated from the initial mass of the Pu-241 in the fuel, assuming that the Pu-241 was two years old since the last time the Am-241 was separated from the plutonium. The Am-241 generated during irradiation will be calculated from neutron captures by Pu-239 and Pu-240. The initial masses of Pu-239 and Pu-240 will be given by AK.

4.4.8 Np-237

The initial mass of Np-237 in a fuel element will be calculated from the initial mass of Pu-241, assuming that the Pu-241 was two years old since plutonium reprocessing. The generation paths of Np-237 will be calculated from neutron captures by U-235 and U-236, and (n,2n) reaction on U-238. The initial masses of U-235, U-236 (zero if not given), and U-238 will be given by AK.

4.5 Supplemental Information for Actinides Inventory Calculation

The calculation of the inventory of the special actinides requires initial values of several isotopes not normally reported in an AK package. In addition, the generation of the special actinides depends on the reaction cross-sections at locations in the EBR-II reactor where the fuel elements were irradiated. These cross-sections have not been developed for an AK package. The following two subsections discuss how the initial values of certain actinides and the cross-sections will be defined for use in the RH TRU waste characterization.

4.5.1 Initial Heavy Metal Masses

Fuel element inventory quantification of Pu-238, Pu-241, Pu-242, Am-241, and Np-237 requires the initial mass values of these actinides, which are generally not obtainable from AK information. The experimental fuel elements irradiated in the EBR-II reactor derived their plutonium from plutonium production reactors. As an approximation to estimating the isotopic ratios from irradiation in the production reactors, average isotopic ratios from irradiation in a pressurized water reactor and a heavy water will be used for the isotopic composition of plutonium.⁵ The ratio of Pu-240 to Pu-239 in a fuel element is given by AK, so the contents of the other plutonium isotopes can be obtained from the Pu-240 to Pu-239 ratio. The Am-241 content will be deduced from the decay of Pu-241 and the Np-237 content will be deduced from the decay of Pu-241 to Am-241 and Am-241 to Np-237.

4.5.2 Reaction Cross-sections

The calculation of the generation of the special actinides resulting from irradiation requires reaction cross-sections. These reaction cross-sections varied across the core of the EBR-II reactor, particularly at subassemblies of different fuel designs and along the fuel elements in the axial direction. In the EBR-II reactor, a fast reactor, most of the neutron flux is in the 100 keV and the 1 MeV energy range, where the (n, γ) and the (n,f) cross-sections do not vary rapidly. The (n,2n) and (n,3n) cross-sections vary rapidly with energy in this energy range, but their values are more than three orders of magnitude below the (n, γ) and (n,f) cross-sections. Consequently, the amount of the special nuclides generated through the (n,2n) and (n,3n) reactions are many orders of magnitude smaller than those generated through the (n, γ) reactions, so large uncertainties in the (n,2n) and (n,3n) reaction cross-sections can be tolerated without much impact on estimating the total special actinide activities. For this reason, detailed reactor models will not be used to calculate the reaction cross-sections at any specific location on a fuel element, but instead, a uniform cross-section set will be used to calculate the average special actinides inventory in any

fuel element. The inventory concentration at fuel element or at any specific location on a fuel element that became part of the waste stream will deviate from the concentration calculated by using the uniform cross-section set. Such deviations will be considered part of the uncertainty in the inventory.

The INEEL is in the process of obtaining average cross-sections at fuel subassemblies of several designs, including metallic and oxide fuel subassemblies with different numbers of fuel elements per subassembly. An average cross-section set computed from these cross-section sets will be used in the special actinides calculations. The variations of the cross-sections from the average will be considered in the uncertainty analysis. The characterization program, however, is not contingent upon the requisition of these cross-sections. As shown in the "Special Actinides" document,⁵ cross-sections averaged from three ORIGEN2 fast reactor libraries^{8,9} give results quite close to (higher by about 30%) results calculated by using detailed reactor models for all the special actinides except U-233. In the case of U-233, if the ORIGEN2 (n,2n) and (n,3n) cross-sections are adjusted upwards by a factor of 2.5, the U-233 results will match the results of detailed reactor model calculations. In the absence of the specific EBR-II cross-sections, these generic cross-sections, when adjusted upwards by a factor of 2.5 for the (n,2n) and (n,3n) reactions, will be used to calculate the special actinide inventory. The uncertainties in the results will increase, but they will not impact the total or TRU activity by more than 20%.

4.6 Confirmatory Calculation

The methods to calculate the Sr-90 and Cs-137 activities in the irradiated fuel elements are straightforward and the accuracies of the results only depend on the accuracies of parameters obtainable from AK. The methods to calculate the special actinides inventory, however, are approximations that give overestimates of the quantities of the special actinides (only generation and no depletion) by a fraction on the order of the burnup (approximately 10%). In addition, the approximations ignore minor reaction paths and intermediate products of short half-lives in the generation of these isotopes. To assess the adequacy of the approximations, ORIGEN2⁸ calculations will be performed using the same cross-section set that is used in the approximate calculations. The ORIGEN2 calculations will include all reaction paths and also depletion calculations for the isotopes. The ORIGEN2 calculations must reproduce approximately (within 20%) the burnup of the heavy metal isotopes (U-235, U-238, Pu-239, and Pu-240) documented in the AK package. The results of the ORIGEN2 calculations will be compared to the results of the approximate calculations. If any value for the special actinides calculated by ORIGEN2 exceeds that calculated by the approximate method, then the ORIGEN2 value will replace the approximate value. Calculation of the uncertainties of the expected values, however, will still be based on the approximate formulation.

5. REQUIRED RADIOLOGICAL PARAMETERS

In Section 4, we presented methods to estimate the masses of the 10 reportable radionuclides, (i.e., Am-241, Pu-238, Pu-239, Pu-240, Pu-242, U-233, U-234, U-238, Sr-90, and Cs-137). In addition, methods to estimate the Pu-241, Np-237, Ba-137M, and Y-90 quantities are also presented. In this section, we present methods that use the masses or activities of these isotopes to calculate the required radiological parameters for irradiated fuel elements and the radiological parameters for a batch of RH TRU waste, which is a fraction of the total inventories of these parameters in the fuel elements that contributed to the RH TRU waste.

5.1 Total Activity

The sum of the activities of the following isotopes, in approximate order of descending activity, comprises over 95% of the total activity in any irradiated fuel element at times approximately 13 years after irradiation:

Pu-241, Cs-137, Ba-137M, Sr-90, Y-90, Pu-239, Pu-240, Am-241, and Pu-238.

Once the time of characterization is selected, the quantities of the radionuclides, Pu-241, Cs-137 and daughter Ba-137M, Sr-90 and daughter Y-90, and Pu-238 determined for the time at the end of irradiation will be decay-corrected to the time of characterization, and the quantities of Am-241 and Np-237 will be adjusted for Pu-241 decay. Because of the long lives of the transuranic isotopes (Pu-239 and Pu-240), these isotopes need not be decay-corrected.

To account for the activities of the isotopes not specifically listed, the total activity will be computed as 1/0.95 of the sum of the activities of the above isotopes.

5.2 Ten Reportable Radionuclides

The 10 reportable radionuclides, Am-241, Pu-238, Pu-239, Pu-240, Pu-242, U-233, U-234, U-238, Sr-90, and Cs-137 will be reported by both mass (g) and activity (Ci) for the time of characterization. U-235, Np-237, and Pu-241 will also be reported as supplemental radionuclides.

5.3 TRU Activity

Over 99% of the total TRU activity comes from Pu-238, Pu-239, Pu-240, and Am-241. The sum of their activities and those of Pu-242 and Np-237 will be reported as the total TRU activity.

5.4 Highest-Hazard Radionuclides

The highest-hazard radionuclides that comprise over 95% of the total radioactive hazard of a fuel element, i.e., Pu-239, Am-241, Pu-240, Pu-241, and Pu-238, will be reported both in curies and in hazard index.

5.5 Activity Concentration

The average activity concentration in a batch of waste is the total activity in the batch divided by the total mass of the waste. The activity concentration in the waste in a waste drum, however, depends on how the total activity is distributed among the drums in a batch. The method for apportionment to

individual drums is discussed in Section 6.2 in conjunction with uncertainty estimates at the waste drum level.

5.6 Dose Rate Determination

Dose rates around waste drums were measured at the time of packaging the waste at ANL-E. Such information can be obtained from the AK information for the waste. However, the dose rates determined at the time of packaging are not average dose rates around the drums, but are rather maximum dose rates around the drums measured at selected elevations of the drums. For the reporting of dose rates, measurements under an accepted Quality Assurance Program will be made before shipping the waste to WIPP. These measurements will also be used for allocating total activities to individual drums and for calculating the total activity concentrations of waste in individual drums.

6. WASTE GENERATION FROM DESTRUCTIVE EXAMINATION OF FUEL ELEMENTS

At the AGHCF, the fuel elements were sectioned into examination samples and the samples were ground and polished for metallographic examinations. Based on ANL-E AGHCF operating procedures, 1.5% of irradiated fuel in a sectioned fuel element was considered to be cleaned up as RH TRU waste. The 1.5% will be applied to the inventory in the fuel elements to obtain the activities in the RH TRU waste.

7. UNCERTAINTY ESTIMATES

The radiological parameters determined for the RH TRU waste that originated from the ANL-E AGHCF will be either based on values documented directly in AK or calculated based on AK and supplemental information. These values will be either best estimates (most likely values) or conservative estimates (values higher than actual). The only conservative estimate that may decrease the confidence of meeting the waste acceptance criteria is the TRU concentration in the waste, which could place non-TRU waste (less than 100 nCi/g) into the TRU category. However, this will not pose a practical problem, because concentration limit for TRU designation is many times the estimates for any batch of the RH TRU waste.

For each radiological parameter of the RH waste, uncertainties will be estimated for the value of the parameter determined for the waste. Uncertainties will be estimated at both the batch level and the waste drum level. The methods for determining these uncertainties are described in the following subsections.

7.1 Uncertainty Estimates at the Batch Level

The basic parameters determining the radioisotope inventory in a batch of waste are: (a) the radioisotope inventory in the fuel elements that contributed to the waste; and (b) the fraction of the radioisotope inventory in the fuel elements that ended up in the waste. The radioisotope inventory in the fuel elements was the sum of the radioisotope inventories of the individual fuel elements. The radioisotope inventory in an individual fuel element depends on material composition in the unirradiated fuel element, burnup of the fuel element, and the time interval since the irradiation of the fuel element. The fraction of a fuel element that ended up as RH TRU waste depended on the destructive examination process, the programmatic requirements for the experiments, such as the number of fuel segments to be prepared and examined, and the waste management practices at the AGHCF.

The method of computing the uncertainty in the radioisotope contents in a batch of waste is to estimate the uncertainty at every step in the process of generating the waste and then to combine the uncertainties to arrive at an uncertainty for the batch. The uncertainty in the radioisotope inventory concentration in part of a fuel element that became RH TRU waste is computed from the uncertainty in its burnup relative to the average burnup of that fuel element, whose average inventory concentration is a function of the initial composition, average burnup, fission yields for fission products, actinide generation rates, and time after irradiation. The uncertainties in all these parameters will be considered except for the time after irradiation, which is accurately documented in the AK. The uncertainty for the batch is then computed from the uncertainties in the inventory of the individual fuel elements. In computing the composite uncertainties (for a fuel element or a batch), the correlation between the various parameters determining the radioisotope inventory will be taken into consideration. If the constituent parameters of a composite parameter are correlated (e.g., fission yields for each fuel element), then their uncertainties (standard deviations) are added to obtain the uncertainty for the composite parameter. If the constituent parameters are uncorrelated, then the variances (squares of the uncertainties) of these parameters are added to obtain the variance of the composite parameter. Partially correlated uncertainties will be treated as correlated uncertainties so that a larger (conservative) composite uncertainty is obtained.

The best-estimate value for the radioisotope inventory in a batch of RH TRU waste will be computed as a constant fraction (1.5%) of the radioisotope inventory of the examined fuel elements during the period the waste batch was consolidated. Uncertainties in this average fraction will be considered first at the fuel element level and then at the batch level (many fuel elements combined). Uncertainties in batch mixing will also be considered. (Here batch mixing refers to accounting the waste

in a consolidation batch from a fuel element examined during another consolidation batch period, or periods, or vice versa.)

7.2 Uncertainty Estimates at the Waste Drum Level

The variation of radioisotope contents at the waste drum level (variations from drum to drum in a batch) will be determined by apportioning the radioisotope contents determined for the batch to the individual drums based on results of dose rate measurements.

The dose rate measurements for a drum will be used to calculate the Cs-137 content in that drum. If the sum of the calculated Cs-137 contents in the drums in the batch falls within the uncertainty band (one sigma) of the AK-based Cs-137 contents, the fission product contents in the drums will be apportioned from the fission product contents in the batch based on the total AK-based Cs-137 contents for the drums. The uncertainty in the fission product content in a drum will be calculated from the uncertainty in the total fission product content for the batch and the uncertainty in the dose rate measurements (standard deviation of several measurements). If the sum of the dose rate based Cs-137 contents falls outside the uncertainty band of the AK-based Cs-137 contents, the discrepancies will be resolved by further investigations, such as gamma spectroscopic measurements and review of the AK documents. These investigations will be case-specific because the reasons for the discrepancies cannot be anticipated.

The TRU and other actinides contents in a drum are generally not in direct proportion to the fission products in the drum because of the fuel elements may have very different initial composition and burnup (from less than 1% to more than 15%). To apportion the TRU and the other actinides to individual drums based on dose rate requires the ratios of the isotope activities to the Cs-137 activity. These ratios will be calculated for the batch, but their variations from fuel element to fuel element will be considered as the uncertainties in the ratios. The uncertainties in the actinides contents in a drum are then calculated from their uncertainties in the batch, uncertainties in the isotopic ratios, and uncertainties in the dose rate measurements.

8. REFERENCES

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Appendix A
Radionuclide Contents for Batch of RH TRU
Waste Drums 728–737

Appendix A

Radionuclide Contents for Batch of RH TRU Waste Drums 728–737

This appendix contains the results of calculations for the radionuclide contents in RH TRU waste drums 728-737 based on the methodology presented in the main body of this report. The radionuclide contents are reported at both the batch level and the drum level. Uncertainties at the drum level are considerably higher than at those at the batch level because of dose rate measurement uncertainties and actinide to Cs-137 ratio uncertainties. Apportionment to drums is based on the average of the recorded dose rates at the time of shipment of the drums from ANL-E to the INEEL. The dose rates for a drum were measured at five regions around the drum. The recorded dose rates were maximum dose rates for each region. Therefore, such dose rates cannot be used to estimate the Cs-137 contents in the drums, but are useful only in apportioning the radionuclides from the batch total to the drums.

All uncertainties are quoted at the one-sigma level.

A.1 Batch Level Parameters

A.1.1 Total activity (uncertainty) – 13.52 Ci (6.23 Ci).

A.1.2 Activities (uncertainties) of ten reportable radionuclides.

Isotope	Expected Value (g)	Uncertainty (g)	Expected Value (Ci)	Uncertainty (Ci)
Am-241	5.69E-02	2.49E-02	1.95E-01	8.56E-02
Pu-238	2.85E-03	1.59E-03	4.88E-02	2.72E-02
Pu-239	3.83E+00	1.26E-01	2.38E-01	7.83E-03
Pu-240	6.51E-01	1.95E-02	1.48E-01	4.44E-03
Pu-242	1.02E-02	2.72E-03	3.88E-05	1.04E-05
U-233	3.39E-06	1.60E-06	3.28E-08	1.55E-08
U-234	7.05E-02	2.08E-02	4.41E-04	1.30E-04
U-238	8.19E+00	1.25E-01	2.75E-06	4.22E-08
Sr-90	1.20E-02	1.45E-03	1.63E+00	1.97E-01
Cs-137	2.67E-02	2.22E-03	2.32E+00	1.93E-01

A.1.3 TRU activity (uncertainty) – 0.630 Ci (0.296 Ci); TRU activity concentration (uncertainty) – 2,708 nCi/g (1,273 nCi/g).

A.1.4 Highest-hazard radionuclides the comprise at least 95% of the total hazard. Maximum allowable hazard index for transportation is 3000. The total hazard (uncertainty) is 134 (65).

Isotope	Activity (Ci)	Uncertainty (Ci)	Hazard Index	Hazard Index Uncertainty
Pu-239	2.38E-01	7.83E-03	4.40E+01	1.45E+00
Am-241	1.95E-01	8.56E-02	3.61E+01	1.58E+01
Pu-240	1.48E-01	4.44E-03	2.74E+01	8.21E-01
Pu-241	4.44E+00	1.87E+00	1.64E+01	6.93E+00
Pu-238	4.88E-02	2.72E-02	9.02E+00	5.04E+00

A.1.5 Activity concentration (uncertainty) – 58.1 μ Ci/g (26.8 μ Ci/g), or 11.9 mCi/L (5.5 mCi/L).

A.2 Drum Level Parameters

A.2.1 Total activity.

Drum No.	Total Activity (Ci)	Total Activity Uncertainty (Ci)
728	0.94	0.37
729	0.52	0.21
730	1.29	0.66
731	2.31	2.10
732	0.60	0.46
733	0.14	0.07
734	4.23	3.91
735	0.08	0.02
736	2.39	1.63
737	1.01	0.52

A.2.2 Activities of reportable radionuclides in mCi.

Drum No.	Am-241	Uncertainty	Pu-238	Uncertainty	Pu-239	Uncertainty	Pu-240	Uncertainty	Pu-242	Uncertainty
728	1.36E+01	2.62E+01	3.39E+00	6.48E+00	1.65E+01	3.63E+01	1.03E+01	2.15E+01	2.69E-03	4.34E-03
729	7.58E+00	1.46E+01	1.89E+00	3.62E+00	9.23E+00	2.03E+01	5.76E+00	1.20E+01	1.50E-03	2.43E-03
730	1.87E+01	3.65E+01	4.66E+00	9.04E+00	2.27E+01	5.04E+01	1.42E+01	2.99E+01	3.70E-03	6.09E-03
731	3.34E+01	7.00E+01	8.34E+00	1.73E+01	4.06E+01	9.52E+01	2.54E+01	5.68E+01	6.62E-03	1.20E-02
732	8.71E+00	1.77E+01	2.18E+00	4.39E+00	1.06E+01	2.43E+01	6.62E+00	1.44E+01	1.73E-03	3.01E-03
733	2.04E+00	3.99E+00	5.09E-01	9.88E-01	2.48E+00	5.51E+00	1.55E+00	3.27E+00	4.04E-04	6.67E-04
734	6.11E+01	1.29E+02	1.53E+01	3.18E+01	7.44E+01	1.75E+02	4.64E+01	1.04E+02	1.21E-02	2.20E-02
735	1.19E+00	2.27E+00	2.97E-01	5.60E-01	1.45E+00	3.14E+00	9.03E-01	1.86E+00	2.36E-04	3.74E-04
736	3.45E+01	6.93E+01	8.62E+00	1.71E+01	4.20E+01	9.51E+01	2.62E+01	5.65E+01	6.85E-03	1.17E-02
737	1.46E+01	2.86E+01	3.65E+00	7.07E+00	1.78E+01	3.94E+01	1.11E+01	2.34E+01	2.90E-03	4.77E-03
Drum No.	U-233	Uncertainty	U-234	Uncertainty	U-238	Uncertainty	Sr-90	Uncertainty	Cs-137	Uncertainty
728	2.28E-06	1.77E-06	3.06E-02	7.18E-02	1.91E-04	3.94E-04	1.13E+02	6.39E+01	1.61E+02	8.71E+01
729	1.27E-06	9.96E-07	1.71E-02	4.01E-02	1.07E-04	2.20E-04	6.33E+01	3.62E+01	9.01E+01	4.94E+01
730	3.13E-06	2.64E-06	4.21E-02	9.96E-02	2.63E-04	5.49E-04	1.56E+02	1.01E+02	2.22E+02	1.40E+02
731	5.60E-06	6.33E-06	7.53E-02	1.87E-01	4.70E-04	1.04E-03	2.79E+02	2.78E+02	3.97E+02	3.90E+02
732	1.46E-06	1.49E-06	1.96E-02	4.78E-02	1.23E-04	2.65E-04	7.28E+01	6.28E+01	1.04E+02	8.78E+01
733	3.42E-07	2.91E-07	4.59E-03	1.09E-02	2.87E-05	6.00E-05	1.70E+01	1.13E+01	2.42E+01	1.55E+01
734	1.03E-05	1.17E-05	1.38E-01	3.43E-01	8.61E-04	1.92E-03	5.10E+02	5.15E+02	7.26E+02	7.24E+02
735	1.99E-07	1.44E-07	2.68E-03	6.23E-03	1.67E-05	3.41E-05	9.92E+00	4.79E+00	1.41E+01	6.43E+00
736	5.79E-06	5.53E-06	7.78E-02	1.87E-01	4.86E-04	1.04E-03	2.88E+02	2.28E+02	4.10E+02	3.18E+02
737	2.45E-06	2.07E-06	3.29E-02	7.79E-02	2.06E-04	4.29E-04	1.22E+02	7.99E+01	1.73E+02	1.10E+02

A.2.3 TRU activity.

Drum No.	Activity (mCi)	Uncertainty (mCi)	TRU Concentration	
			Mean (nCi/g)	Uncertainty (nCi/g)
728	43.8	50.0	2,682.1	3,063.6
729	24.5	28.0	2,073.5	2,370.9
730	60.2	69.6	3,588.2	4,148.4
731	107.7	132.2	6,086.2	7,471.1
732	28.1	33.7	1,998.6	2,392.5
733	6.6	7.6	402.3	465.9
734	197.1	242.7	7,758.9	9,550.9
735	3.8	4.3	103.0	116.5
736	111.3	131.6	2,610.7	3,086.1
737	47.1	54.5	1,365.7	1,580.0

A.2.4 Hazard index of the highest-hazard radionuclides that comprise over 95% of the total hazard. Maximum allowable hazard index for transportation is 300.

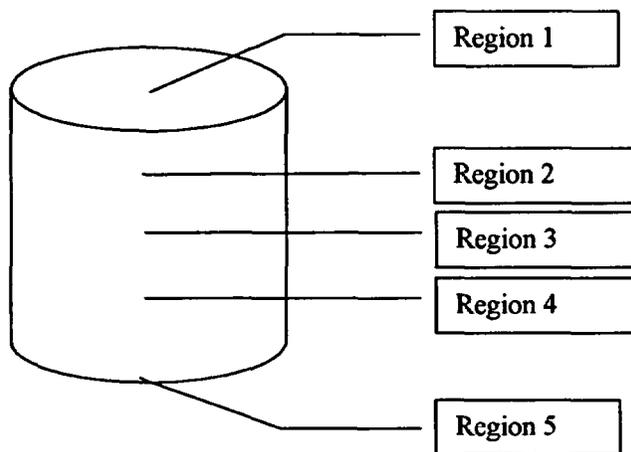
Drum No.	Pu-239		Am-241		Pu-240		Pu-241		Pu-238	
	Activity	Uncertainty								
728	3.06E+00	6.70E+00	2.51E+00	4.84E+00	1.91E+00	3.97E+00	1.14E+00	2.24E+00	6.27E-01	1.20E+00
729	1.71E+00	3.74E+00	1.40E+00	2.70E+00	1.06E+00	2.22E+00	6.38E-01	1.25E+00	3.50E-01	6.69E-01
730	4.20E+00	9.21E+00	3.45E+00	6.66E+00	2.62E+00	5.46E+00	1.57E+00	3.08E+00	8.62E-01	1.65E+00
731	7.51E+00	1.65E+01	6.17E+00	1.19E+01	4.69E+00	9.76E+00	2.81E+00	5.50E+00	1.54E+00	2.94E+00
732	1.96E+00	4.30E+00	1.61E+00	3.11E+00	1.22E+00	2.55E+00	7.34E-01	1.44E+00	4.02E-01	7.68E-01
733	4.58E-01	1.01E+00	3.76E-01	7.26E-01	2.86E-01	5.96E-01	1.71E-01	3.36E-01	9.40E-02	1.80E-01
734	1.37E+01	3.02E+01	1.13E+01	2.18E+01	8.58E+00	1.79E+01	5.14E+00	1.01E+01	2.82E+00	5.39E+00
735	2.67E-01	5.86E-01	2.20E-01	4.24E-01	1.67E-01	3.47E-01	1.00E-01	1.96E-01	5.48E-02	1.05E-01
736	7.77E+00	1.70E+01	6.38E+00	1.23E+01	4.85E+00	1.01E+01	2.91E+00	5.69E+00	1.59E+00	3.04E+00
737	3.28E+00	7.20E+00	2.70E+00	5.21E+00	2.05E+00	4.27E+00	1.23E+00	2.41E+00	6.74E-01	1.29E+00

A.2.5 Activity concentration.

Drum No.	Total Activity Concentration			Total Activity Concentration	
	Activity (Ci)	(μ Ci/g)	Concentration (μ Ci/g)	Concentration (mCi/L)	Uncertainty (mCi/L)
728	0.94	57.5	22.6	8.3	3.3
729	0.52	44.5	18.0	4.6	1.9
730	1.29	76.9	39.3	11.4	5.8
731	2.31	130.5	118.8	20.3	18.5
732	0.60	42.9	32.7	5.3	4.1
733	0.14	8.6	4.5	1.2	0.7
734	4.23	166.4	154.0	37.2	34.5
735	0.08	2.2	0.6	0.7	0.2
736	2.39	56.0	38.1	21.0	14.3
737	1.01	29.3	15.1	8.9	4.6

A.2.6 Surface dose rates of drums.

The surface dose rates shown here are the maximum dose rates measured in five regions around a drum at the time when the drums were prepared for shipment to the INEEL from ANL-E in 1992. The different regions are shown in the following figure.



Drum No.	Ave Dose Rate (R/hr)	Uncertainty (%)	Region 1 Dose Rate (R/hr)				
728	2.4	30.9	2.5	2.5	3.5	2	1.5
729	1.3	32.4	0.9	1.5	1.9	1.5	0.9
730	3.3	44.9	4	4	5	2	1.5
731	5.9	87.7	2.5	3	4	5	15
732	1.5	72.4	0.8	1.25	3.5	1.25	0.9
733	0.4	46.5	0.5	0.5	0.4	0.3	0.1
734	10.8	89.3	10	27	10	4.5	2.5
735	0.2	10.6	0.2	0.25	0.2	0.2	0.2
736	6.1	63.6	4.5	8	12	3	3
737	2.6	45.6	3	4	3	2	0.9