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LOCKHEED MARTIN

DEVELOPMENT OF THE REMOTE-HANDLED TRANSURANIC WASTE RADIOASSAY DATA QUALITY OBJECTIVES

An Evaluation of RH-TRU Waste Inventories, Characteristics, Radioassay Methods and Capabilities

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LOCKHEED MARTIN ENERGY RESEARCH CORPORATION
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

ORNL-27 (3-96)

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Date Published - September 1997

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U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-84OR21400

EXECUTIVE SUMMARY

This report results from a task implemented in the fall of 1996 to assist the National Transuranic Program Office of the Department of Energy/ Carlsbad Area Office (DOE/CAO) in establishing data quality objectives (DQOs) for the radioassay of Remote-Handled Transuranic (RH-TRU) waste. As part of the task a draft report was submitted to DOE/CAO on Jan. 10, 1997. This report is being published as an ORNL Technical Memo to provide a referenceable document, to add information not included as part of the draft report previously submitted to DOE/CAO, and to incorporate comments from the RH-TRU sites throughout the complex.

The DQO process is a methodical, seven-step approach for balancing decision uncertainty with available resources [EPA-1, DOE-EM-1]. Consistent with the DQO process, this report identifies regulatory drivers and practical issues of RH-TRU management; consolidates site-specific information on current RH-TRU management practices and plans, particularly as they apply to the radioassay of transuranic radionuclides; discusses the use of process knowledge and its role in RH-TRU radioassay; identifies previous, existing, and near-existing capabilities for radioassay; and proposes development needs that will allow for the final development of DQOs and corresponding Quality Assurance Objectives. This report pertains to radioassay issues only.

This report is a compendium of information resulting from many sources, including discussions and visits with knowledgeable personnel at several DOE sites, the review of radioassay data submitted in the Baseline Inventory Report, rev. 3 [CAO-8]; and evaluation of radioanalytical methods used throughout the complex for measuring and reporting radionuclides.

The RH-TRU waste generator sites discussed in this report are: Argonne National Laboratory-West, Los Alamos National Laboratory, Idaho National Engineering and Environmental Laboratory and Oak Ridge National Laboratory. The DOE installations at Hanford and Battelle Columbus are not discussed because of the uncertainty into which their category waste will be managed. Additionally, expert elicitation was sought for regulatory-driven issues related to the WIPP performance assessment and the RH-TRU 72B shipping cask.

The required quality of RH-TRU radioanalytical measurements is driven by two regulatory issues: transportation, and the repository performance assessment (PA). The 72B shipping cask must be approved by the Nuclear Regulatory Commission. The Environmental Protection Agency must be assured that WIPP will be operated in compliance with 40CFR. Of the two drivers, the NRC will most likely play a larger role in the approval process for RH-TRU waste because the impact of RH-TRU waste on the performance assessment is negligible, as reported in the Compliance Certification Application (CCA) for 40CFR191. [CAO-7] DQOs and associated QAOs should be formulated to satisfy two requirements, in order of priority:

- Transportation parameters, as delineated in the 72B-cask Safety Analysis Report, shall not be exceeded. In most cases, the most influential parameter is ^{239}Pu fissile gram equivalent (fge); in fewer cases, the influential parameter may be thermal power. In yet other cases, operational constraints (e.g. high dose rates) may actually be the most limiting design parameter.

- The relative abundance of radionuclides shall be reported at a sufficient quality level that enables WIPP to a) demonstrate that the PA-modeled, relative isotopic inventory used to originally demonstrate compliance with 40CFR191 [CAO-7] continues to be valid as actual RH-TRU waste is moved into the WIPP facility over time; or b) demonstrate compliance with 40CFR191 via mandatory 5-year re-executions of the PA model using actual reported data. [CAO-1]

Radioassay is accomplished by the resourceful combination of process knowledge, sampling and radiochemical analysis, and bulk measurement by nondestructive assay. For a considerable fraction of RH-TRU waste, process knowledge will play a larger role than for the CH-waste counterpart. For instance, special nuclear material accountability and control and computations of fuel burnup and reactor fission-product inventory will provide the backbone of process knowledge for RH-TRU waste originating from within hot cells. For sludge waste, process knowledge of the chemical and radionuclide content of the waste will provide an envelope for describing waste material composition. Radiochemistry and bulk radioassay must be combined more effectively for RH-waste. Proficiency of non-destructive assay (NDA) on real RH-TRU debris waste has been demonstrated in one case at LANL. NDA proficiency for other real RH waste streams has not been demonstrated and is not well known. On the other hand, proficiency of destructive assay (DA) on aged (8-10 years) RH-TRU waste is well known and not significantly different from CH waste. The primary limitation of RH-TRU waste characterization by DA is sampling, therefore applying rigorous quality assurance objectives to measurements adds little value. Development efforts for RH-TRU NDA are in early stages. Additional efforts are needed to make NDA techniques a useful application for RH-TRU radioassay. DA methods are fairly mature but are not standardized for RH-TRU waste characterization.

DQOs must be formulated with essentially one endpoint in mind: the use of radioassay needs to be performed at a quality level that assures compliance with transportation and waste acceptance criteria. This does not necessarily mean that a radioassay measurement must be made within a few percent, but rather, that when the uncertainty of the radioassay is added to the mean, the resulting value does not exceed an acceptable limit. Inherent in this logic is that the closer a generator packs a waste shipment to the limit, the better his measurement accuracy should be. Otherwise he can choose to manage his waste shipments in a way so as to optimize the process. There is no choice but to develop implementable DQOs that balance risk, cost, and available resources; this balance is termed “risk benefit”. For RH-TRU waste, DQOs should be established in such a way as to provide latitude to generators in actually satisfying them. Within this document is a clear description of the regulatory drivers, waste generator inventories and descriptions of the waste and existing and near existing radioassay capabilities. Also included in the document (specifically section 5) are recommendations for continuing the DQO process for RH-TRU waste.

ACKNOWLEDGEMENTS

The authors wish to express appreciation to the following staff members of the ORNL RH-TRU Radioassay DQO Development Team who made important contributions to this work:

J. M. Keller
T. H. Monk
F. J. Schultz
J. R. Stokely

The authors also wish to express appreciation to staff members working in waste management, non-destructive assay laboratories, and radiochemical laboratories at the following DOE sites for participation in the site visits.

Argonne National Laboratory-West
Idaho National Engineering & Environmental Laboratory
Los Alamos National Laboratory
Oak Ridge National Laboratory
Sandia National Laboratory (contribution to performance assessment evaluation)
Westinghouse (72B Cask NRC-license submittal)

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ACRONYMS

AMWTF	Advanced Mixed Waste Treatment Facility
ANL	Argonne National Laboratory
ANL-E	Argonne National Laboratory-East (Chicago, IL)
ANL-W	Argonne National Laboratory-West (Idaho Falls, ID)
APNEA	Active-Passive Neutron Examination and Assay
ASTM	American Standard Test Methods
AWCC	Active Well Coincidence Counter
BCL	Battelle Columbus Laboratory (Columbus OH)
BVEST	Bethel Valley Evaporator Service Tanks (Oak Ridge)
CAO	Carlsbad Area Office
CCA	Compliance Certification Application
CCDF	Complimentary Cumulative Distribution Function
CFR	Code of Federal Regulation
CH	Contact Handled
CH-TRU	Contact-Handled Transuranic
CMR	Ceramic Metals Research Facility
COC	Certificate of Compliance
CTEN	Combined Thermal Epithermal Neutron
DA	Destructive Assay
DDT	Differential Dieaway Technique
DOE	Department of Energy
DOE-EM	Department of Energy Environmental Management
DOT	Department of Transportation
DQO	Data Quality Objective
EBR	Experimental Breeder Reactor
EML	Environmental Measurements Laboratory
EPA	Environmental Protection Agency
ER	Environmental Restoration
FATS	Fission Assay Timing System
FGE	Fissile Gram Equivalent
GAAT	Gunite and Associated Tanks (Oak Ridge)
GNAT	Gamma Neutron Assay Technique
HFEF	Hot Fuel Examination Facility (Argonne-west)
IAEA	International Atomic Energy Agency
ICPP	Idaho Chemical Processing Plant
INEEL	Idaho National Engineering and Environmental Laboratory
INMM	Institute of Nuclear Materials Management
IPAN	Imaging Passive Active Neutron

LANL	Los Alamos National Laboratory
LCS	Laboratory Control Sample
LLLW	Liquid Low Level Waste
MAPEP	Multi-analyte Performance Evaluation Program
MCNP	Monte Carlo Neutral Particle
MVST	Melton Valley Storage Tanks (Oak Ridge)
NDA	NonDestructive Assay
NDA/NDE IWG	NonDestructive Assay/NonDestructive Examination Interface Working Group
NRC	Nuclear Regulatory Commission
NTPO	National Transuranic Program Office
NTS	Nevada Test Site
OHF	Old Hydrofracture Facility
OHFT	Old Hydrofracture Facility Tanks
ORIGEN	Oak Ridge Isotope GENeration
ORNL	Oak Ridge National Laboratory
PA	Performance Assessment
PAN	Passive Active Neutron
PIC	Passive Institutional Control
PK	Process Knowledge
PNCC	Passive Neutron Coincidence Counting
PNL	Portsmouth National Laboratory
QA	Quality Assurance
QAPP	Quality Assurance Program Plan
QAPjP	Quality Assurance Project Plan
QAO	Quality Assurance Objective
QC	Quality Control
RCRA	Resource Conservation and Recovery Act
REDC	Radiochemical Engineering Development Center (Oak Ridge)
RH	Remote Handled
RH-TRU	Remote-Handled Transuranic
RTF	Remote Treatment Facility (Argonne-west)
S&A	Sampling and Analysis
SARP	Safety Analysis Report
SNL	Sandia National Laboratories
SGS	Segmented Gamma Scanner
SNM	Spent Nuclear Material
TRU	Transuranic
TWBID	Transuranic Waste Baseline Inventory Database
TWBIR	Transuranic Waste Baseline Inventory Report
WAC	Waste Acceptance Criteria

WF	Weighting Factor
WUF	Waste Unit Factor
WIPP	Waste Isolation Pilot Plant

1.0 INTRODUCTION

1.1 Scope and Purpose

The Waste Isolation Pilot Plant will accept remote-handled transuranic waste as early as October of 2001. Several tasks must be accomplished to meet this schedule, one of which is the development of Data Quality Objectives (DQOs) and corresponding Quality Assurance Objectives (QAOs) for the assay of radioisotopes in RH-TRU waste. DQOs, as defined by the Environmental Protection Agency are:

> "Qualitative and quantitative statements derived from the DQO process that clarify study objectives, define the appropriate type of data, and specify the tolerable levels of potential decision errors that will be used as the basis for establishing the quality and quantity of data needed to support decisions." [EPA-1]

The Department of Energy requires that the DQO process be instituted for all significant data collection projects within the Office of Environmental Management to provide a basis for balancing decision uncertainty with available resources. Figure 1.1 illustrates the concepts of the DQO process for waste characterization. The process is an adaption of the seven step EPA DQO process to waste characterization. [DOE-EM-1]

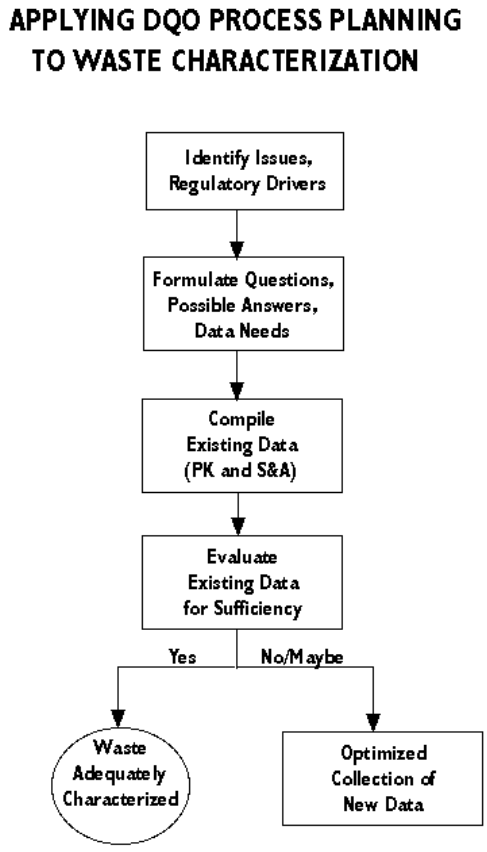


Figure 1.1 DQO Process Planning

Oak Ridge National Laboratory (ORNL) was assigned the task of providing to the DOE CAO, information necessary to aide in the development of DQOs for the radioassay of RH-TRU waste. Consistent with the DQO process, information needed and presented in this report includes:

- identification of RH-TRU generator site radionuclide data that may have potential significance to the performance of the WIPP repository or transportation requirements;
- evaluation of existing methods to measure the identified isotopic and quantitative radionuclide data;
- evaluation of existing data as a function of site waste streams using documented site information on fuel burnup, radiosotope processing and reprocessing, special research and development activities, measurement collection efforts, and acceptable knowledge; and

- the current status of technologies and capabilities at site facilities for the identification and assay of radionuclides in RH-TRU waste streams.

This report is intended to provide guidance in developing the RH-TRU waste radioassay DQOs, first by establishing a baseline from which to work, second, by identifying needs to fill in the gaps between what is known and achievable today and that which will be required before DQOs can be formulated, and third, by recommending measures that should be taken to assure that the DQOs in fact balance risk and cost with an achievable degree of certainty.

Limitations of the Report

The information in this report was obtained through review of applicable requirements documents, review of the Transuranic Waste Baseline Inventory Report [CAO-8], consultation with the RH-TRU generator sites, and correspondence with a panel of radiochemical and nuclear measurement experts. The scope of this report is limited by the ability to consolidate an abundance of radioassay information obtained via generator site visits and correspondence. It is meant to provide an overview of the site generator information as it is applicable to the development of DQOs for radioassay. Not all RH-TRU generator sites and waste streams are represented in this report nor is a full detailed evaluation provided, but applicable references are cited. Generator sites which are represented are identified in Section 4.

1.2 Background

The WIPP facility, scheduled to open in May 1998, is operated by the DOE to demonstrate the safe disposal of TRU waste produced as a result of national defense activities. TRU waste, according to the DOE Order 5820.2A , is radioactive waste that contains greater than 100 nanocuries per gram of alpha-emitting isotopes with atomic numbers greater than 92 and half lives greater than 20 years. [DOE-1] The two categories of TRU waste are contact-handled, with external radiation dose rates measuring no greater than 200 mrem/hr at the surface of a waste container, and remote-handled, with radiation dose rates measuring greater than 200 mrem/h but less than 1000 rem/hr at the surface of the waste container. By public law 102-579, no more than 5% of the waste volume going into WIPP can have contact dose rates in the range between 100 rem/h and 1000 rem/h.

1.3 Establishing RH-TRU Radioassay Data Quality Objectives

Acceptance of transuranic waste for disposal at the WIPP requires chemical, radiological, and physical characterization of the waste for completion of WIPP permit applications and certification in accordance with the WIPP waste acceptance criteria. [CAO-4] The guidelines for characterizing contact-handled transuranic (CH-TRU) waste are incorporated into the *Transuranic Waste Characterization Quality Assurance Program Plan (QAPP)* [CAO-5]; however, the current edition of this document does not contain DQOs and guidelines for characterizing RH-TRU waste. Thus, for RH-TRU waste, either the existing QAPP needs to be amended or a separate QAPP for RH-TRU waste needs to be written in a manner that accounts for the differences in the management of RH and CH waste. Section 2 of this report explains why the DQOs for RH waste are not

expected to parallel the DQOs for CH waste; the governing concerns and issues are entirely different. For RH-TRU waste, a much greater emphasis needs to be placed on assuring that individual transportation requirements are not exceeded. Section 2 essentially represents the first box illustrated in figure 1.1. Section 3 corresponds well with the second block of figure 1.1. The third and fourth boxes are addressed by section 4 which presents a compilation of existing data from four RH-TRU generator sites: Argonne National Laboratory-West, Los Alamos National Laboratory, Idaho National Engineering and Environmental Laboratory and Oak Ridge National Laboratory. The report concludes in section 5 by summarizing how each of the DQO planning steps were addressed with the report and identifies additional activities needed to complete the DQO process.

2.0 WASTE CERTIFICATION REQUIREMENTS

2.1 RH-TRU Waste Acceptance Criteria for Radioassay

To accomplish the tasks of characterizing, certifying, packaging, transporting, and dispositioning remote-handled transuranic (RH-TRU) waste at WIPP requires the development of a Quality Assurance Program Plan (QAPP) to assure the regulators and the public, with some level of confidence, that the Waste Acceptance Criteria are met. Waste is certified when it can be demonstrated that the waste acceptance criteria have been satisfied. For RH waste, the applicable criteria that depend on radioassay are summarized in table 2.1. [CAO-4, Table 3.7]

These acceptance criteria depend on radioassay in the following way: the value of each parameter is derived from the measurement of constituent isotopes, followed by a normalized summation over all isotopes. While it has been shown that thermal power and ^{239}Pu fissile gram equivalent (fge) are directly measurable parameters---by calorimetry and active-neutron measurements, respectively--- the remaining two parameters rely on the measurement of individual isotopes.

Table 2.1 Radioassay Dependent, RH-TRU Waste Acceptance Criteria

Parameter	Limit
Administrative subcritical mass limit (^{239}Pu fissile gram equivalent, *fge*)	≤ 600 g/canister < 325 g/cask
^{239}Pu Equivalent Inhalation-Dose Activity	≤ 1000 PE-Ci/ canister
TRU Alpha Activity	> 100 nCi/g ≤ 23 Ci/litre
Thermal Power	< 300 watt/canister

For example, ^{239}Pu-equivalent activity is calculated by the following expression:

$$^{239}Pu_{eq} = \sum_{i=1}^{n} \frac{A_i}{WF_i} \qquad [\,Ci\,]$$

where: A_i = *Activity of radionuclide, i* [*Ci*]

WF_i = *Weighting Factor of radionuclide, i*

Where the weighting factor for each i^{th} isotope is given by the ratio:

$$WF_i = \frac{H_{239Pu}}{H_i}$$

where: H_{239Pu} = *committed eff. whole body dose eq. per* μCi, ^{239}Pu
H_i = *committed eff. whole body dose eq. per* μCi, *nuclide i*

TRU alpha activity is computed in a similar way, where the summation of activity (A_i) includes each isotope that meets the TRU definition. Fissile gram equivalent and thermal power are the only two criteria that may be arrived at by either direct measurement or by summation over the constituent isotopes. It should be noted that when a criteria can be determined by direct measurement it is more defensible that calculated results. This helps with the confusion brought about by summing over "all" constituent isotopes and not knowing when to say a sufficient number of isotopes have been determined. For RH-TRU waste this becomes extremely important as illustrated by example below.

What is important to note for these waste acceptance parameters is that they are impacted minimally, <u>if at all</u>, by the large presence of fission and activation products found in RH waste. That is to say, the sensitivity of the waste acceptance parameters to large inventories of non-transuranic radionuclides in RH waste is insignificant. For example, the ^{239}Pu equivalent activity parameter increases by the same amount for every curie of ^{239}Pu or for every 16,000 curies of ^{137}Cs[1]. Another example is the consequence of ^{90}Sr on the fge parameter; it is zero because ^{90}Sr is not fissile. These are very important points related to RH waste because the largest fraction of radioactivity in RH waste is from isotopes that do not impact any of the radioassay-dependent waste acceptance criteria, particularly relative to the influence of transuranic radioisotopes. This fact will later be formulated as a recommendation for performance/risk-based DQOs (see §5).

2.2 Regulatory Drivers for RH-TRU Waste

The origin of these waste acceptance criteria lies primarily within two regulatory-based efforts to ensure that the waste can be stored for 10,000 years with acceptable risk to people and that it can be transported safely across public highways:

a) WIPP Performance Assessment (PA) and Compliance Certification Application (CCA) to demonstrate that the EPA requirements of 40CFR191.13(a) are satisfied; and

[1] The committed effective whole body dose equivalent per unit microcurie inhaled (assuming a 1-micron activity median aerodynamic diameter) for ^{239}Pu is 510 mrem(W) while for ^{137}Cs it is 0.032 mrem (D). The inhalation dose equivalent ratio is thus 16,000. [DOE-2]

b) Safety Analysis Report (SARP) for the 72B shipping cask and the certificate of compliance to be issued by the Nuclear Regulatory Commission in the 1997 to 1998 time frame.

The ensuing discussion will explain the rationale for emphasizing the transportation-based driver over the performance assessment-based driver in establishing data quality objectives for RH-TRU waste.

2.2.1 Repository Performance Assessment

In October of 1996, the Department of Energy submitted to EPA the "Title 40 CFR 191 Compliance Certification Application for the Waste Isolation Pilot Plant." This submittal included the latest results from the Sandia National Laboratory (SNL) performance assessment computational-modelling team. These latest efforts account for the most important isotopes governing the PA [SAND-1, SAND-4], and provide results for both undisturbed and disturbed performance, including a sensitivity analysis for total radionuclide release from the repository. Latest results utilized the radionuclide data as reported in the Transuranic Waste Baseline Inventory Report, rev. 3. [CAO-8]

In the undisturbed performance case, results show compliance with individual-human and groundwater protection requirements. In the bounding approach to the dose calculation, the maximum annual committed effective dose to a receptor is 0.47 mrem. The allowable limit is 15 mrem.

Performance, in the disturbed case, also meets EPA standards for compliance. The largest impact to repository performance is from human intrusion, specifically those human activities associated with direct releases to the surface: cuttings, cavings, and spallings.

The results of the latest analyses show that the following EPA requirements of 40CFR191.13(a) will be met:

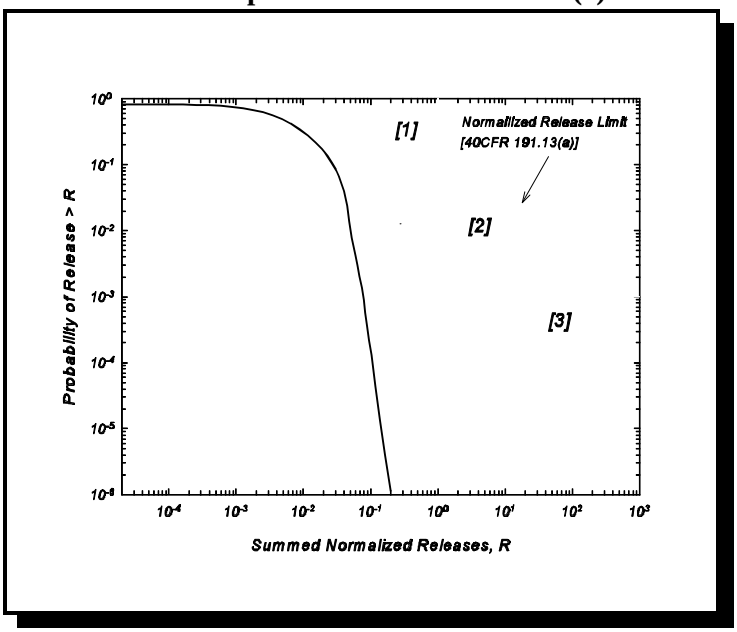
> "Disposal systems for spent nuclear fuel or high-level or transuranic radioactive wastes shall be designed to provide a reasonable expectation, based upon performance assessments, that the cumulative releases of radionuclides to the accessible environment for 10,000 years after disposal from all significant processes and events that may affect the disposal system shall:
>
> (1) Have a likelihood of less than one chance in 10 of exceeding the quantities calculated according to Table 1 (§191, appendix A); and
>
> (2) Have a likelihood of less than one chance in 1,000 of exceeding ten times the quantities calculated according to Table 1 (§191, appendix A)."

Demonstrating compliance graphically, the probability of exceeding the normalized sum of a release, R, is plotted against the release, R, as shown in figure 2.1. The maximum allowable release limit is

shown by the indicated boundary to the right. Expressing the release as a "normalized" release[2] accounts for isotope-specific release limits for containment requirements listed in table 1 of appendix A, 40§191.

As seen in region [1] of figure 2.1, it is acceptable for the probability of a normalized release of less than 1 to be greater than 1 in 10. Similarly, region [2] shows that the probability of incurring a normalized release greater than 1, but less than 10 times a unit-normal release, can be no larger than 1 in 10 and no smaller than 1 in 1000. Finally, region [3] shows that the probability of exceeding 10 times a unit-normal release must be less than 1 in 1000. Compliance with the EPA requirements in 191.13(a) is achieved when the CCDF does not extend to the right of the limit lines. The CCDF plotted in figure 2.1 is for illustrative purposes only, but is nearly consistent graphically with most recent data provided by SNL. Figure 2.1 shows that the most recent data suggests that WIPP performs better than the EPA requirements by at least a factor of 10, for the entire projected inventory of both CH and RH waste. Aside from potential errors unbeknownst to the modeling team at this time, the only major impact to these results would be caused by significant changes in relative isotopic composition. That is to say, performance of the repository is not impacted by the magnitude of the activity (because it's normalized by the Waste Unit Factor (WUF)), but rather, is impacted only when reported isotopic ratios significantly

Figure 2.1. Illustration of Complimentary Cumulative Distribution Function (CCDF) for showing compliance with 40CFR191.13(a).



[2]There are three key radionuclide inventory parameters which contribute to the "normalized" release values; the 40CFR191 Waste Unit Factor ((WUF) the number of allowable alpha-curies that may be stored in the repository), the allowed radionuclide releases from Table 1 of Appendix A 40 CFR191, and the EPA Unit. The WUF is computed by adding the inventory (in the year 2033, post closure) of all alpha-emitting TRU radionuclides with half-lives > 20 years and dividing by 10^6. SAND-5 reports the total alpha activity at closure as 3.44E+06 Ci and hence a resulting WUF of 3.44 (See Appendix A). To arrive at a release limit per isotope, appendix A 40§191 limits per WUF are multiplied by the WUF of 3.44. To demonstrate compliance with 40§191, the "Source EPA Unit" per radionuclide is defined as the radionuclide inventory divided by the allowable release. The total Source EPA Unit is then determined by summing over all alpha emitters, yielding in this case 1.00E+04 [SAND-5]. This is interpreted as "WIPP may release 1 ten thousandth of its inventory over 10,000 years on a unit normalized basis". As the "real" inventory diverges from the "projected inventory, the WUF may change by some multiple and not affect the outcome of meeting the performance objectives on the basis of a normalized release. It is when the relative isotopic abundances change to a degree that then affects the currently computed 1.0E+04 Source EPA Unit.

change beyond those which were reported in the BIR, rev. 3. Once WIPP is operational, the impact of changes to the "real" reported inventory is evaluated by mandatory re-execution of the PA model. This is required every 5 years.

For establishing DQOs on RH waste, a relevant question to ask is:"what is the performance assessment impact of RH-waste, relative to CH-waste?" The answer is: negligible. For example, table 2.2 shows the volume and total radioactivity fractions for each of the two TRU wastes. RH waste is a small fraction of CH waste in terms of volume and radioactivity. Furthermore, table 2.3 shows the isotopes corresponding to 94% of the radioactivity for RH waste and 97% of the radioactivity for CH waste, accounting for almost all the radioactivity reported in table 2.2, 14% and 86%, respectively. [CAO-8]

Table 2.2 Relative Influence of CH and RH Waste on WIPP Capacity [CAO-8]

	Remote Handled	Contact Handled	Total
Volume			
cubic	$7{,}000 m^3$	$168{,}500 m^3$	$175{,}500 m^3$
percent	4%	96%	
Activity			
curies	1×10^6	6×10^6	7×10^6
percent	14%	86%	

Table 2.3 Significant Isotopic Differences Between RH and CH TRU Waste

This table presents the primary radionuclides dominating the RH and CH TRU inventories based upon activity.

Remote Handled			Contact Handled		
Isotope	**Percent of Total RH Activity**	**Percent of Total TRU Activity**	**Isotope**	**Percent of Total CH Activity**	**Percent of Total TRU Activity**
^{137m}Ba	20%	3%	^{238}Pu*	40%	35%
^{137}Cs	20%	3%	^{241}Pu	36%	31%
^{90}Sr	20%	3%	^{239}Pu*	12%	11%
^{90}Y	20%	3%	^{241}Am*	6%	6%
^{241}Pu	14%	2%	^{240}Pu*	3%	3%
Total	94%	14%		97%	86%

*TRU Isotopes

There are a few relevant points to make about Table 2.3:

(a) The largest fraction of radioactivity in CH waste is from transuranic radionuclides. Including ^{241}Pu, which is not TRU, but decays relatively quickly to the TRU radionuclide ^{241}Am, nearly 100% of the CH activity is comprised of TRU radionuclides. WAC parameters are the most sensitive to these radionuclides.

(b) The largest fraction of radioactivity in RH waste is from fission products. WAC parameters are least sensitive to these radionuclides as described in §2.1.

(c) RH radioactivity is relatively small. The only radionuclide that "produces" a TRU-waste radionuclide (^{241}Pu to ^{241}Am) comprises only 2% of the total repository inventory, on a radioactivity basis. Even if the RH radionuclide ratios change appreciably the impact on a unit normalized release will be minimal relative to the CH influence (see footnote 2).

(d) What the table doesn't reveal are the results reported by Sanchez in his SNL memo of April 25, 1996 [SAND-4] and in the WIPP PA Analysis Report, EPAUNI, [SAND-5]. SNL identified the important radionuclides to model in the PA: first, the four radionuclides of ^{238}Pu, ^{239}Pu, ^{241}Am, and ^{240}Pu account for 98.83% of the EPA unit release. Virtually all of this radioactivity is found in CH waste. Because of the importance of these results relative to the DQO basis for RH waste, the summary tables from SAND-5 are provided in appendix A of this report. The first table in App. A shows the 5 top radionuclides as cumulative percentages to the EPA unit. The second table breaks table 1 down further into CH and RH components. From these tables, it is quite clear that the RH component has at least a factor of 10 and often a factor of 100 less influence on repository performance than CH waste.

(e) To even make a stronger case that RH-waste is relatively inconsequential to the long term performance of WIPP, it appears that EPA will allow credit for Passive Institutional Controls (PICs) up to 700 years, during which time the radioactivity of RH-waste on a specific volume basis will be far less than the CH counterpart.[3]

In summary, PA/CCA-based regulatory drivers should not influence DQOs for the radioassay of RH-TRU waste. The fact that the volume and activity fractions of RH-TRU waste are so small relative to CH waste and the fact that the radioactivity of RH waste is largely comprised of short-lived fission products forces one to look elsewhere for regulatory-based drivers that affect prudent decision making.

2.2.2 Transportation

On December 20, 1996, the Department of Energy submitted to the Nuclear Regulatory Commission the Safety Analysis Report [WEST-1] for the 72B RH-TRU shipping cask, requesting from the NRC that a Certificate of Compliance (COC) be issued. The NRC approval is in progress and upon approval will confirm the RH-TRU-specific data quality specifications. Additional impact to the regulation of RH-TRU shipments may result from the promulgation of new DOT regulations, to be

[3] Private communication with L. Sanchez, Sandia National Laboratories.

consistent with the International Atomic Energy Agency's HM169A. [IAEA-1] What can be stated for now is the following: radioactive material transportation requirements have been regulated on the basis that the shipper has in place, acceptable measurement methods, to assure that the shipment is categorized, packaged, labeled, and placarded correctly and that the shipping manifest is adequately completed to allow emergency response personnel to make well-informed decisions in the event of an emergency involving a collision and/or accidental release[4]. Therefore, the primary transportation-based driver appears to be that which assures the individual radioassay-based parameters approved in the NRC COC are not exceeded.

For transportation, there are five primary safety parameters:

1) the Bill of Lading must be completed accurately enough to assure that in the event of an accident, response personnel are able to make well-informed decisions on mitigation efforts. While the DOT regulations have changed recently and are still in the process of change to comply with HM-169A, it is common practice to report all radioisotopes in the shipment that comprise 99% of the activity and those that comprise 95% of the hazard. The authors are not aware of any quality assurance standards established for this determination. Logically, the level of quality should be sufficient to ensure that no gross errors are made in handling the material that would lead to significant health consequences either during accident situations or under normal operation, that otherwise would have been avoided had "better" radioassay been achieved. The TRU program should look for precedence in other shipping programs such as for experimental fuel residue, spent nuclear fuel, or low-level radioactive waste;

2) subcritical mass limits for a shipment shall not be exceeded. This parameter provides assurances that under any circumstance encountered during transport, the shipment will remain subcritical;

3) heat generation rate for a shipment shall not be exceeded. This parameter provides assurances that the maximum engineered heat load and gas-generation rates are not exceeded;

4) radiation dose rates at the surface of the cask and at all other boundaries as defined by DOT shall not be exceeded. This parameter protects personnel, the driver, and the public from exposure to ionizing radiation. This parameter is always measured prior to shipment. If the shipment exceeds limits, it is reconfigured;

5) ^{239}Pu equivalent activity for a shipment shall not be exceeded. This parameter assures that in the event of a release, the maximally exposed individual from inhalation of 1-micron sized particles

[4] There is only one case known to the authors whereby the NRC questioned the use of scaling factors for estimating radionuclide content in low-level waste originating from NRC-regulated nuclear facilities. See IE INFORMATION NOTICE NO. 86-20: Low Level Radioactive Waste Scaling Factors, 10CFRPart61

is below regulatory thresholds. The fundamental basis, i.e. decision point, for this parameter is not known by the authors.

2.2.3 Summary of Regulatory Drivers

DQOs and QAOs for CH-TRU waste were established from a viewpoint dealing with the impact of the waste on repository performance. TRU alpha activity was chosen as the most suitable DQO. QAOs were established with a two-fold emphasis: 1) to discriminate low-level waste from TRU waste at the 100 nCi g^{-1} fiducial and 2) to partition the alpha-activity ranges into four ranges based on the largest existing fraction of CH-TRU in storage at the INEEL. These decisions were well founded.

If the same rationale is applied to the RH-TRU case, the results will be different because the regulatory emphasis for RH-TRU waste is quite different from CH-TRU waste. The regulatory-drivers from transportation are relatively more imposing, particularly the parameters fissile gram equivalent and thermal power. On this basis, a suitable DQO for RH-TRU waste is not TRU alpha activity, but rather fissile gram equivalent. TRU alpha activity should be deleted as a DQO for RH-TRU waste. Also, there is no reason to divide QAOs into 4 levels, as was done for the CH-TRU case. It is apparent, based on our review, that the most important objective of radioassay is to assure that WAC limits are not exceeded (table 2.1). For RH-waste the risk of placing waste less than 100 nCi/g in the repository is far less than the implications of exceeding WAC parameters.

Based on an evaluation of regulatory drivers, it is of primary interest is to assure that individual shipment criteria are not exceeded. Of secondary interest is to assure that the relative abundance of radioisotopes is known well enough to provide “reasonable” data to future executions of the PA models.

3.0 RADIOASSAY METHODOLOGIES AND APPLICATION TO RH-TRU WASTE

Radiochemistry is the science of measuring the quantity of radionuclides; in practice the scope includes the separation chemistry, measurement, and detection of radiation. Radiochemistry is often referred to as radioassay. Radioassay provides the ability to determine the presence of radioisotopes and their quantities in sampled materials. To perform radioassay with emphasis on risk-benefit, process knowledge plays an important role for three reasons: to focus the investigation on the radionuclides of interest for a specific application; to establish boundary conditions on the radioassay effort; and to provide estimates of radionuclides that otherwise are difficult to directly measure. Radioassay can be performed on materials, in bulk quantity, or on samples collected from the bulk material; referred to as nondestructive assay, and destructive assay, respectively. This section introduces the concepts of radioassay, tailored to RH-waste, recognizing that a risk-effective and timely approach to performing the task of radioassay is accomplished by balancing resources against data quality objectives. It is very important on the front end to assure that data quality objectives are realistically established to avoid unnecessary risks on the back end. Radioassay is capable of providing very accurate results, provided a commensurate level of resources are provided to accommodate the particular level of rigor required.

For nearly 70 years, a large amount of knowledge and expertise has been amassed on radioassay. For contact-handled waste, sampling and analysis plans for destructive measurements have been written and approved throughout the DOE. Nondestructive assay instruments have been used effectively for measuring radioisotopes in both product and waste streams. These methods can be adapted to the particular challenges facing the radioassay of RH-TRU waste. It is clear, however, that a more balanced approach between the use of process knowledge, destructive assay, and nondestructive assay will have to be used for RH-TRU waste more than has been used for assay of CH-TRU waste, [CAO-5].

3.1 Process Knowledge

The concept of process knowledge and its utilization are embodied in the WIPP RCRA Part B Permit Application, appendix C9, as acceptable knowledge [CAO-6]. In CAO-6, the role of acceptable knowledge is described in great detail that will not be reported here, not only because of its length, but more importantly, its application to radioassay is different from that of RCRA characterization.

Some widely used forms of process knowledge for RH-TRU waste include reactor fuel burn up calculations, analysis of the product stream, sampling and analysis of the initial waste stream, and waste generator records.

Fuel burn-up and ORIGEN codes

As described in §4.2, debris waste from hot cell facilities at LANL, ANL-E, and ANL-W consists of irradiated experimental fuel fragments, fines, and turnings. Calculations (and some measurements) can be made to

establish fission and activation products generation in the fuel by reactor irradiation. Results from experimental fuel-burnup calculations are normally combined with results from calculations of fission and activation production to provide very accurate radionuclide inventory distributions on an individual fuel-pin basis. The most widely used code for computing fission- and activation- product yield in reactor fuel is ORIGEN (Oak Ridge Isotope GENeration). Because it is so widely used, and the fact that ORIGEN data appears to be generally available and accepted, a brief overview on its utility to RH-TRU as process knowledge is described in Appendix B.

Sampling and Analysis of Initial Waste Streams

Sampling and analysis of an initial waste stream can provide an entire profile of a waste stream, such as the information obtained from sampling and analysis of ORNL sludge (see §4.5.4), or it can provide information on the relative isotopic abundance or spatial distribution of a waste stream. The type of sample collected depends upon the type of information needed. Obtaining an entire profile of a waste stream requires a representative sampling of the waste stream with complete analysis of the sample. Combining initial waste stream sampling and analysis with a well defined and controlled process for waste treatment and packaging can virtually eliminate the need to characterize individual packages of the final waste form. Only confirmation of the sampling and analysis information would be warranted by random sampling and analysis or random screening with non-destructive assay techniques.

Another common form of sampling and analysis process knowledge involves a selective sampling of the waste stream using smears or actual samples of the waste to obtain relative isotopic abundances of the stream. This type of process knowledge can be very useful for debris waste in which representative samples are impossible to obtain without otherwise shredding and homogenizing the material. Relative isotopic abundances can be combined with dose rates and assumptions regarding the volume contamination of the waste to arrive at a rough estimation of the high gamma-ray flux radionuclide content for the entire waste package. Isotopic distributions are also useful in determining scaling factors to associate hard-to-measure radionuclides to direct NDA measurements. An example of using this relation to quantify bulk quantities of waste is performing a direct measurement of the bulk waste, such as gamma spectrometry, and applying the scaling factors determined from process knowledge to calculate transuranic nuclides and fission products[5].

Waste Generator Records

Waste generator records are typically required of all generated waste across DOE sites in the form of logbook entries or waste stream profile forms. The amount of information included in the records varies depending upon the waste management requirements implemented during the time of waste generation. Information provided in waste generator records may include descriptions of the waste, the process from which the waste was generated, product information from the process, dose rates, and qualitative estimations of the primary

[5] This has been directly attempted at ORNL, but for waste where the TRU activity <<<< non-TRU activity, the estimate of TRU activity has very large error bands. One of the difficulties is that “hot spots” of activity, if sampled, will significantly bias the isotopic proportions. Additional biases are introduced by schemes used to chemically isolate the isotopes of interest for radioassay. [ORNL-16]

radionuclides present. Although waste generator records often provide considerable information, it is difficult to relate the information to the radionuclide content of actual waste items. In most cases, waste generator records are used in addition to other forms of acceptable knowledge for making conclusions about the waste radionuclide content.

For RH-TRU waste, process knowledge provides, in particular, information before radioassay, that will prove vital for:

- demonstrating the waste to be transuranic and not spent fuel or high level waste;
- establishing an envelope of possible radionuclides to analyze;
- establishing a baseline from which to plan the approach to radioassay;
- determining isotopic content of difficult-to-measure radionuclides by scaling results from direct NDA measurement;
- providing direction for making effective use of resources for sampling; and
- prioritizing the list of potential effects that must be calibrated for during radioassay measurements.

3.2 Non-destructive Assay

Nondestructive assay (NDA) measures penetrating radiation emitted from containerized radioactive material. Detected radiation is related to the radionuclides present and their quantities. It is convenient, rapid, and in many cases can provide an accurate measure of radioactivity packaged in containers from 1 to 3- gallons in size up to drums of 30- to 55- gallons in size. Some systems have been designed to accommodate B25-box sized containers.

On the one hand, NDA is widely used because of its appeal in reducing sample collection of hazardous materials. Because NDA is a completely non-intrusive measurement, it obviates the need for chemical separation of isotopes from one another; material processing to reduce radiation dose rates to levels manageable by the analytical laboratory; and management of hazardous/radioactive waste materials generated by destructive analysis. As a result, exposure of personnel to radiation and hazardous substances is greatly reduced. For applications in which NDA is applicable, the sampling error that otherwise is associated with sampling of the material, is negligible. Because bulk measurement by NDA describes the average radioactivity of the entire container, multiple NDA measurements of the same container are not required for improving the quality of the radioactivity estimate.

On the other hand, NDA is not a panacea; there are restrictions and it can suffer significantly from matrix effects in large containers. There are many cases where NDA, under given field conditions, simply cannot measure radioisotopes that are present. Hence, NDA results often must be combined with destructive analyses and process knowledge. This allows one to scale unmeasured isotopes to measured isotopes, to make better corrections for matrix and source effects, and to adjust parameters of the measurement system to achieve an optimized response. Unlike DA, where the sample is prepared in a manner to minimize these interferences, NDA methods must be evaluated to determine the effect of interferences on a given measurement. This can

give rise to large corrections and is normally not a simple task when the nominal accuracy desired is in the 20% range for measurements of 55-gal sized containers of heterogeneous TRU waste. Measurement accuracy is improved when the waste stream is consistent (from drum to drum), smaller containers are assayed, radiation rates are "nominal", the radioactive material is not clumped together, and important isotopes are not masked by less important, but more intense isotopes.

3.2.1 NDA Theory

Nondestructive assay is essentially a three step process: measure radiation, associate the radiation with a specific radionuclide (or radionuclides), and then determine the amount of each radionuclide. The first step is measuring the radiation, which has to be penetrating enough to get out of the package being radioassayed. Figure 3.1 is an illustration of the physical phenomenon. Alpha particles (α) deposit all of their energy in the package. None penetrate the skin of the package. Beta particles (β) deposit most of their energy in the package. The fraction of energy that escapes from the package is normally of such low intensity that it is not useful. In containers with walls that are fairly thick and are of high atomic number (e.g. iron), beta particle interactions can produce x-rays from Bremmstrahlung, depicted as B-xray. These too are normally of no use to NDA. The two types of radiation that do emerge from the package with sufficient intensity to be analytically useful are gamma-rays (γ) and neutrons (n). The only "arrows" in figure 3.1 that cross the container boundary are those from neutrons and gamma-rays. Gamma-rays have a nice feature: they can identify exactly the presence of a specific isotope. Nearly all isotopes emit a given set of discrete-energy gamma-rays, commonly called a gamma-ray signature. Neutrons do not have this feature, but are very useful for measuring transuranic radionuclides because TRU radionuclides either emit neutrons during decay, or may be fissioned, to produce more neutrons. The other type of radiation depicted in figure 3.1 is heat. Heat emitted from a package of radioactive material can be measured and related to the radioactivity content. The NDA methods that make use of these physical phenomena are normally grouped into three categories: gamma-ray based spectroscopy, neutron counting, and calorimetry. NDA methods are non-intrusive and non-invasive, thereby providing benefits in the management of these hazardous materials.

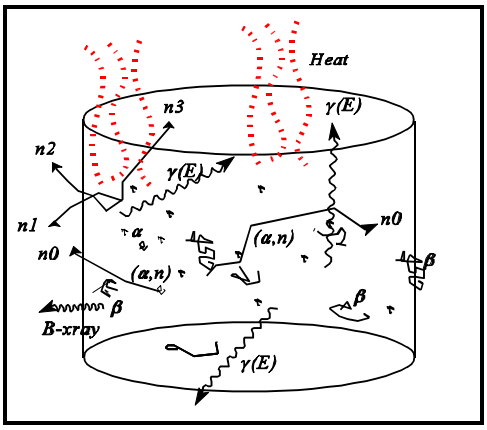


Figure 3.1 Illustration of Radiation Emission from a Waste Container

Gamma-ray methods are referred to as spectroscopic because the individual gamma-ray energies can be resolved by direct measurement, thereby providing information about a specific isotope. Corrections for matrix interferences are made by "shining" a known gamma-ray source through the container. This is called an active or transmission-source correction. Measurements are normally made in vertical slices, hence the term segmented gamma-ray scanning. Latest technology not only

slices vertically, but also laterally and radially---this is called computed tomography. Correcting for matrix interferences in large containers is very important. When small amounts of radioactivity are present, gamma-ray methods require long count times (several hours). When Ci-quantities of radioactivity are present, detectors become overwhelmed and need to be shielded and collimated. Gamma-ray interactions in the waste matrix and within the detector lead to the detection of compton-scattered photons which impede sensitivity. Inter-related effects are determined and corrected for during the calibration phase.

Neutron methods best measure two categories of radioisotopes: fissile and those that spontaneously fission. They also measure all alpha-emitters indirectly, by detecting the neutrons emitted from alpha capture on lightweight nuclei like fluorine and oxygen---but this feature turns out normally to be more of a hindrance than a help. Fissile isotopes are measured actively. Spontaneously-fissioning isotopes are measured passively. Active neutron methods use an external neutron source to irradiate the container. Differential-dieaway uses active neutron to measure ^{239}Pu and ^{235}U. Active-well coincidence counters and ^{252}Cf shufflers utilize the same principle but accomplish it quite differently. Passive neutron methods rely on the detection of two (or more) neutrons in coincidence with one another, i.e. arriving within the same time. The time-coincidence neutron rate is related to the number of spontaneous fissions that took place, and thus to the amount of ^{240}Pu, ^{242}Pu, and ^{252}Cf. Unlike gamma-ray methods, neutron measurements cannot estimate the activity of individual isotopes. Process knowledge and/or measures of isotopic proportions must be used to partition out the contributions to the signal from each constituent isotope. Active neutron measurements provide a direct WIPP parameter, fissile gram equivalent (FGE).

Calorimetry is in theory, a straightforward technique. There are many different types of calorimeters, but the most widely used type of radiometric calorimeter is of the heat flow/isothermal variety because the heat rate from radioactive decay is essentially constant over the measurement period. [NUREG-1] Calorimetry does not measure any single isotope specifically, but rather, the heat output of the container. A radiometric calorimeter consists of a chamber that is immersed in a bath at some constant temperature. The bath can be air or water, for example. An electrical circuit, whose resistance is temperature dependent, is utilized to measure very small changes in temperature. Calorimeters used for small samples can measure in the 100 uW range. Measurement times are proportional to desired sensitivity and the time for the temperature between the sample and the chamber to equilibrate. The alternative to measuring heat generation, is to indirectly compute heat generation by quantifying the contributing isotopes in a waste package and then sum the individual contributions.

3.2.2 NDA Techniques

A general description of the most widely used NDA methods for CH-TRU waste is presented in Appendix C, and there are many references on the subject. [NUREG-1, NUREG-2, ANLW-1] NUREG-1 provides an excellent description of passive NDA methods; NUREG-2 provides an excellent description of active NDA methods. National standards for use of the methods have been

written. [ANSI-1, ASTM-1 through ASTM-5] The adaptation of these methods to RH-waste began about eight years ago. LANL built a system to measure 1-gal buckets of waste within a hot cell. The system was built, used to measure a portion of the LANL RH-TRU waste, and dismantled in 1994. [LANL-1, LANL-2] Other efforts to measure RH-TRU waste using NDA are in development at ANL-W [ANLW-2, ANLW-3], ORNL [ORNL-4, 5,10], INEEL [INEEL-3], as described in §4.4.2.

The most widely used CH-TRU assay systems are Segmented Gamma-ray Scanning (SGS), Passive Active Neutron (PAN); and Passive Neutron Coincidence Counting (PNCC). There are variations on the theme, but the basic physics issues are the same. The following paragraphs describe the challenges posed to these systems for the assay of RH-TRU waste.

Gamma-ray systems configured for CH-TRU waste will have to be redesigned somewhat for application to RH-TRU waste. Most of the design changes will stem from several aspects: shielding and collimating the detector from large gamma-ray fluxes; picking out TRU isotope signatures in the presence of large non-TRU signatures; dealing with dead-time correction; fitting distorted (asymmetric) peaks; and dealing with resolution loss from neutron damage in HPGe detectors. When waste containers are small (1-3 gallons), the challenges of dealing with matrix effects in RH waste will be relatively unimportant. Poor counting statistics at very low TRU activities also become much less important for RH waste.

Present neutron systems were designed to have an optimum neutron detection efficiency both geometrically and intrinsically. This enables these systems to detect very small quantities of fissile material rapidly. This is not the emphasis for RH-TRU waste. Most of the recent publications for the application of neutron systems to CH-TRU waste emphasize how to deal with distortions in the neutron signal as a function of source/matrix position and heterogeneities. [INEEL-1, INEEL-2] When the waste container is kept small (1-3 gallons) for RH-TRU assay, these challenges become less important. When the container is large (30- and 55- gallons), these challenges pose significant problems [ORNL-17, INEEL-6], hence it is in the best interest to keep container size small. What does become important for RH-TRU assay using neutron methods is: 1) the ability to discriminate neutron signal from large-flux/pulse pile up gamma-ray signals, 2) the ability to supply large interrogating neutron fluxes for active-neutron measurements (much greater than 10^8 n s^{-1}) and 3) the ability for the system to recover rapidly from high neutron rates, thus allowing the possibility for the application of high count-rate, multiplicity measurements. Active neutron measurements provide a direct measure of the most important RH-TRU parameter: fissile gram equivalent (fge). Thus, it is very possible that this method could be adapted to ensure that fge is not exceeded in any individual container.

Unlike the case of gamma and neutron systems, the application of calorimeters to date has been mostly on small containers, typically less than a few gallons in size. The application to large gallon size containers needs to be evaluated for feasibility (i.e. the time for equilibration may be several hours to several days and facilities may need significant modification for containment for the calorimetry

system). The application of calorimetry to RH-TRU waste packaged in small containers, may be an excellent, cost-effective approach. This measurement may not assist with the determination of isotopes exclusively, but it does provide a direct measure of an important WAC parameter, thermal loading. This technology could be used to ensure that thermal loading limits are not exceeded.

Implementation of current NDA systems to the RH environment must be proceeded in a manner to account for facility-related issues dealing with high radiation rates. For example, existing maintenance, repair, and development activities for CH-TRU NDA systems is not much of an issue. Radiological controls are minimal. On the other hand, for RH-TRU waste, most of the NDA activities will take place in a hot cell or nearby. Radiological controls for RH-TRU waste will be significant. Hence, all design work needs to be directed toward system simplification. Components must be reliable and easy to replace should problems occur. Accessibility to less reliable components should be optimized. Complicated systems with less reliable parts or many moving subsystems should be avoided. Implementation of systems into the RH environment may actually drive design requirements on the EPA basis of "best available technology" under existing waste management conditions.

3.2.3 NDA Proficiency

The proficiency of NDA techniques for application to RH-TRU waste characterization is difficult to identify at this time due to the lack of standardized techniques, calibration sources, performance evaluation programs and actual application to the various RH-TRU waste streams. The proficiency of CH-TRU NDA systems, on the other hand, is fairly well known:

1. Chapman and Hensley, from ORNL have reported on several occasions [ORNL-6, INEEL-1] the intercomparison of NDA results obtained between measurements of weapons grade (WG) Pu packaged in 55-gal drums made at Nuclear Fuel Services and ORNL. These independent measurements are largely within 30% of each other. In some extreme measurement conditions, results were within a factor of two;
2. Harker, Blackwood, et. al. from INEEL have completed the most comprehensive approach to evaluating total measurement uncertainty by monte carlo uncertainty analysis using Latin Hypercube Sampling. [INEEL-1,2] They showed for graphite molds that the existing PAN configuration would satisfy the QAPP requirements as modified in the latest version. [INEEL-1, CAO-5] For combustible and glass waste, the existing configuration would meet the QAPP requirements at the two middle α activity ranges, but not at the two extremes. [INEEL-2]
3. Prettyman et. al. from LANL and Marts from LLNL have reported on the accuracy of gamma-ray measurements performed at LANL and RFP, and LLNL, respectively. [INEEL-1,2] For WG-Pu in 55-gal drums at activity concentrations greater than several hundred nanocuries/gram, and low density matrices, gamma-ray methods can easily yield results to within 20%. [INEEL-1,2, ASTM-3]

4. Several privatization efforts are underway to demonstrate, by measurement, the performance of developed systems. These tests are being performed at the INEEL. Results were not available at the time of this writing.
5. The most comprehensive testing reported for the performance of PAN systems to measure CH-TRU waste is found in LANL-3. Nicholas, Coop, and Estep report the effects due to matric scattering, absorption, and source self-shielding, the prevailing challenges with the assay of CH-TRU waste.
6. Two cycles of inter-laboratory testing have been completed. These results, which are for nearly "ideal" waste measurement cases, are presented in §3.2.4.

What is currently known about NDA proficiency for RH-TRU waste characterization results from the measurement of hundreds of 1-gallon buckets by LANL and development projects throughout other DOE sites. NDA proficiency for RH waste is summarized below:

- LANL is currently the only DOE site to report NDA results on actual RH-TRU waste. The reported fissile mass results for 13 canisters of LANL RH-TRU waste consisted of an average relative error of 14%. Evaluation of the Passive/Active Neutron (PAN) system and geometries used for these measurements indicates a detection limit of 20 mg fissile material for a benign matrix. This performance is limited to measurements on 1 gallon sized cans, a fairly well known isotopic distribution, a relatively small neutron emission rate, and a known waste matrix with favorable NDA qualities (i.e. not too highly moderating or absorbing). [LANL-1]. Important effects of self shielding were studied and accounted for. No gamma spectrometry measurements were made.

- A project at ANL-W of measuring nine EBR-II assemblies using passive neutron total counting concluded that an indication of total plutonium mass distribution and burnup in blankets, drivers, and ternary assemblies could be obtained. The project results also concluded that the interferences from gamma rays from these highly radioactive assemblies could be overcome through shielding and discrimination. [ANLW-2] This type of data would be useful in confirming process knowledge, but not for characterizing unknown wastes.

- A second project at ANL-W for application of the *Passive Active Neutron* method to the assay of leached cladding hulls (anticipated gamma exposure rates in excess of 100 R h^{-1}; negligible neutron dose rates) showed a measurement accuracy of less than 3% with a precision of less than 4% for passive results. This evaluation was only for determining accuracy of measuring the mass of ^{240}Pu in leached cladding hulls. Active measurements results are not yet available. [ANLW-3]

- INEEL performed an evaluation on the use of Direct Gamma-ray Spectrometry (DGS), Passive/Active Neutron Assay (PAN) and ORIGEN inventory calculations to the characterization of the radionuclide content in their primary RH-TRU waste stream packaged

in 55-gallon drums. The evaluation, based upon Monte Carlo modeling and synthetic gamma-ray spectrum generation, indicated that radionuclide content of their wastes could be determined with relative standard deviations of 20-55%, and detection limits of about 0.06g ^{235}U and 0.04g ^{239}Pu using DGS, PAN, and ORIGEN.

- ORNL has performed research and development studies on the application of NDA to the most difficult RH-TRU waste across the DOE complex, ORNL RH-TRU debris waste with neutron dose rates in the range from 10 mrem h^{-1} to 100's of mrem h^{-1}, and with TRU isotopes accounting for a very small weight and activity fraction of the total radioisotopic mix. Studies of direct fissile mass measurements using a Radiofrequency Quadrupole (RFQ) indicate an estimated detection limit in the 500 nCi g^{-1} range for TRU alpha activity concentration [ORNL-10, ORNL-17] under existing conditions. Studies of measuring actual RH-TRU waste in 3-gallon buckets using the Active Passive Neutron Examination and Assay (APNea) and Segmented Gamma Scanner (SGS) systems have also been used and compared with radiochemistry measurements. The SGS results are within a factor of 4 with the radiochemistry measurements. The results showed that the significant neutron count rate ($>10^6$ neutrons per sec) was too high to be analyzed by the APNea system. More detail on these RH-TRU projects is provided in §4.5.4. Additional work is underway.

All measurement conditions equal, the accuracy of CH-waste NDA measurements will be better than RH-waste measurements by at least 30%. Fortunately, RH systems will not suffer large matrix effects when the waste container is small. But the difficulties associated with high count rate situations will more than offset the gain of not having to deal with matrix effects. Separating signal from noise will degrade sensitivity. The Minimum Detectable Concentration will likely be greater than 100 nCi/g, so this determination will have to be made by other means (i.e. process knowledge and sampling and analysis).

3.2.4 QC Acceptance Criteria

The current version of the *Transuranic Waste Characterization Quality Assurance Program Plan* [CAO-5] section 9 provides quality assurance objectives (QAOs)for the radioassay of CH-TRU waste. The formation of these rules was based upon a review of assay data from 12,205 drums of CH-TRU waste from 50 waste item description codes (i.e. codes used to segregate waste forms based upon physical or tracking differences). The DQO, TRU alpha activity, and the corresponding four levels of activity were derived based on an evaluation similar to that which is reported here for RH waste. The four levels of alpha activity and acceptable ranges for precision, accuracy, and bias are reproduced in table 3.1, directly out of CAO-5, table 9.1.

Table 3.1 Existing Quality Assurance Objectives for the Radioassay of CH-TRU Waste

{Table 9-1, Quality Assurance Objectives, QAPP Interim Change, 11/15/96}

Quality Assurance Objectives

Range of Waste Activity in α-Curies[a]	Nominal Compliance Point α-Curies [a] (g WG Pu)[b]	PARAMETER Precision [c] (%RSD)	Accuracy [d] (%R)	Total Bias[e] (%)	Completeness [f] (%)	MDC (nCi/g) [g]
0	0					60
>0.002 to 0.02	0.008 (0.1)	≤ 20	75-125	Low 25 High 400	100%	
>0.02 to 0.2	0.08 (1.0)	≤ 15	50-150	Low 35 High 300%	100%	
>0.2 to 2.0	0.8 (10)	≤ 10	75-125	Low 67 High 150	100%	
>2.0	12.5 (160)	≤ 5	75-125	Low 67 High 150	100%	

[a] Applicable range of TRU activity in a 208 liter (55 gallon) drum to which the QAOs apply, units are Curies of alpha-emitting TRU isotopes with half-lives greater than 20 years

[b] The nominal activity (of weight of Pu) in the 208 liter (55 gallon) drum used to demonstrate that QAOs can be achieved for the corresponding range in column 1, values in parentheses are the approximate equivalent weights of weapons grade plutonium (WG Pu) , fifteen years after purification; for purposes of demonstrating QAOs, "nominal" means within ± 10 percent

[c] ± one standard deviation based on fifteen replicate measurements of a non-interfering matrix

[d] Ratio of measured to known values based on the average of fifteen replicate measurements of a non-interfering matrix, see Section 9.1.2 of CAO-5 for additional details

[e] 95-percent confidence bounds for system bias established by studies to determine contributions to total uncertainty from all significant sources. Units are confidence bound divided by true value, expressed as a percent. Requirement for the QAO for total uncertainty is to determine and document but no system wide limiting values are established.

[f] Valid radioassay data is required for all waste containers, see Section 9.1.6 of CAO-5 for additional details

[g] As defined in section 9.1 and 9.6 of CAO-5

The ability of a site to meet these QAOs is tested via the Performance Demonstration Program (PDP). The program consists of one to two cycles per year and is required for a site to show compliance with the QAOs for performing NDA of CH-TRU waste. Currently, the PDP is only designed for CH-TRU waste and tests relative precision and accuracy. Total bias is evaluated by other means. [CAO-12] Thus far, two cycles of the PDP have tested NDA systems in 3 matrices of four alpha activity ranges. In all cases, weapons grade plutonium was used as the source of radioactivity: ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am. Under very controlled "blind-test" conditions, six replicates of each of the four configurations were acquired. Table 3.2 summarizes the results of the two cycles (for further details on the plan, current results, and reported technical challenges, refer to CAO-12, CAO-10, CAO-11, INEEL-1, and INEEL-2, respectively).

Table 3.2 NDA PDP Results for Cycles 1 and 2 (CH-TRU waste)

		Neutron-Based Measurements			Gamma-ray Based Measurements		
Matrix	Nominal α Activity (mCi)	No. of Neutron Meas.	**Precision** No. Passed	**Accuracy** No. Passed	No. of Gamma Meas.	**Precision** No. Passed	**Accuracy** No. Passed
Cycle 1 Empty Drum	281	6	6	5	5	4	5
Cycle 1 Ethafoam	490	5	5	5	5	4	5
Cycle 2 Ethafoam	72	6	5	4	5	3	3
Cycle 2 Combustibles	690	7	7	7	5	4	5

Important points related to Table 3.2:

1) level of rigor: these early cycles were intended to be straightforward---modest matrix effects and no source effects, with relatively good count rates. Future test conditions are planned to be progressively more rigorous;
2) CH tests vs. RH test needs: future PDP cycle configurations for CH-TRU waste do not test the conditions appropriate to RH-TRU waste. Tests for CH-TRU waste are projected to evaluate different types of radioactive standards (WG-Pu, RG-Pu, heat-source Pu, HEU, the influence of ^{241}Am ingrowth, and self-shielding) and 55-gal sized matrices (combustible, glass, metal , solidified inorganic-, solidified organic-, and heterogeneous-materials). CH test parameters do not apply to RH-TRU waste, which listed in order of priority are: count rate loss/dead-time correction, pulse pile-up, interference from non-TRU isotopes, and potential self-shielding of the TRU source material. Of much less concern to RH-waste are matrix effects (since large fraction of RH-TRU waste can be assayed in smaller containers), and varying types of TRU standards;
3) scoring criteria: the scoring criteria, to the unseasoned evaluator, can be very misleading. To make the statement that NDA systems perform to the QAPP criteria really means that they perform much better than the upper limits would indicate. First the scoring criteria have been modified to account for only six replicates, versus the fifteen replicates for which Table 3.1 is based. Second, the relative standard deviation parameter is defined relative to the true activity and not the mean of the measurements. While this is statistically correct, it is not practically correct. That is to say, it is possible for a system to exhibit excellent precision, be biased more than 10% relative to the true value and fail the test for precision. Third, and most importantly, the pass/fail decision point is a hypothesis test, with the null hypothesis that the measurement device cannot meet the QAO. As such, the actual measurements must perform well below the upper QAO limits of table 3.1. For example, table A-1 in CAO-12 shows that the QAO of 0.20 (for the smallest

alpha activity range) translates to 0.14 for the noninterfering PDP test and 0.16 for the interfering case. If the test were performed under the null hypothesis that the device can measure to a certain degree of quality, then the output from the independent test could actually slightly exceed the tabled QAO and still satisfy the hypothesis that no statistically-significant data was generated that would lead one to conclude that the device cannot satisfy the QAOs;

4) interfering vs. noninterfering matrix: during the implementation phase of the PDP, QAPP QAOs have been adjusted to reflect acceptable tolerances in the case of an interfering vs. noninterfering matrices. This approach is reasonable for CH-TRU waste on the basis that there is, indeed, a wide range of matrices (contained within 55-gal drums) that do impact the quality of an NDA measurement. This criteria is definitely not the focus in the case of RH-TRU waste; and
5) MDC: there is no plan to test the ability of an instrument to measure TRU alpha activity at the Minimum Detectable Concentration (MDC) at 60 nCi/g. For RH-TRU waste, this QAO is likely to be impossible for most cases; the TRU alpha activity signal is very small relative to the non-TRU signal.

3.3 Destructive Assay

Sampling and analysis is commonly referred to as destructive assay (DA). DA is probably the most widely used method for detecting the presence of radioactivity in a sample collected from the waste. The degree of sensitivity and accuracy achieved with DA is essentially only limited by safety and cost in ensuring a representative sampling scheme is obtained. Therefore, guidelines for DA must be implemented to achieve optimal results with minimal risk. The current version of the *Transuranic Waste Characterization Quality Assurance Program Plan* [CAO-5] section 9 contains quality assurance objectives (QAOs) for nondestructive assay of contact-handled transuranic waste. QAOs for destructive radioassay are not included. While the ability of existing NDA methods to measure TRU radionuclides in RH-TRU waste is greatly impacted by the high radiation background, RH-TRU waste is no more difficult to assay by DA than CH-TRU waste once the sample has been collected and properly handled. This section addresses sampling considerations, identifies analytical methodologies, describes typical instrumentation, and presents feasible quality control criteria for radiochemical analysis of RH-TRU waste. Because the differences between CH-TRU and RH-TRU frequently do not impact laboratory analysis significantly, this section is also applicable to radiochemical analysis of CH-TRU waste.

3.3.1 Sampling

The critical limitation of radiochemical analysis lies in the representativeness of the collected sample. Section 8 of the current version of the *Transuranic Waste Characterization Quality Assurance Program Plan* [CAO-5] provides guidance on obtaining representative samples of “homogeneous” CH-TRU waste via “random sampling” using coring tools to core vertical and horizontal lengths of waste material. This may be applicable to final waste forms like sludge, but certainly not debris waste. Two important points can be made about DA of RH-TRU waste in regards to sampling:

- Sampling of RH-TRU waste (whether sludge or debris) will likely not result in a high level of representativeness, therefore, implementing highly rigorous quality control (QC) acceptance criteria on the radiochemical analyses or other measurements adds little value to the actual quality of the data.

- The risk and cost of collecting RH-TRU waste samples should be factored into the quality assurance objective. [DOE-EM-1, DOE-EM-2, EPA-1]

3.3.2 Radiochemical Methods

A variety of methods are typically used to measure individual radionuclide quantities in a sample collected from the waste. Depending upon the type of sample (solids, smears, etc.) and analysis, sample digestion, dissolution or leaching may be required. Often, additional sample preparation methods are required to isolate certain radionuclides from either the sample matrix or from other interfering radionuclides (i.e. separating pure beta emitters from each other in order to obtain a result for one single beta emitter, such as Sr-90 separation from Y-90). Separation methods play a big role in the quantification of alpha and beta emitting radionuclides in RH-TRU waste. Separation methods typically allow preconcentration of radionuclides from a complex sample matrix. In the case of RH-TRU waste, very small samples may need to be taken and large dilutions performed to decrease personnel exposure to radiation. In addition, certain activities of radionuclides can vary several orders of magnitude within the sample making it difficult to analyze the radionuclides present at low activity ranges. This problem can be rectified in many cases with separation techniques which separate and concentrate the low activity radionuclides from the sample matrix. Due complex RH-TRU waste sample matrices, effective sample preparation methods are needed.

There are few EPA approved methods in the regulatory/compliance arena available for radiological measurements of high-level radioactive wastes. EPA standardized methods for radiological measurements typically involve precipitation methods and numerous reagents and steps which make them time-consuming. These methods do not emphasize reduction in personnel exposure to radiation because they are primarily adapted for environmental level radioactive samples. Numerous radiochemical methods are provided in ASTM literature, but are more directed toward radiochemical processing and other nuclear fuel applications than towards nuclear waste. The only standardized radiological methods which are useful for high level radioactive waste samples include those for sample digestion, gross alpha and gross beta measurements, and gamma spectrometry measurements. These include:

- SW-846 Method 3015, *Microwave Assisted Acid Digestion of Aqueous Samples and Extracts* [EPA-2]

- SW-846 Method 3051, *Microwave Assisted Acid Digestion of Sediments, Sludges, Soils, and Oils*[EPA-2]

- EPA Method 600/900.0, *Gross Alpha and Beta Radioactivity in Drinking Water* [EPA-3]

- EPA Method 600/901.1, *Gamma Emitting Radionuclides in Drinking Water* [EPA-3]

In the early 1990's, an effort to consolidate analytical methods for characterization of DOE waste (including transuranic waste) was started in support of DOE Environmental Restoration Programs. The result of this effort is a living document, the *DOE Methods for Evaluating Environmental and Waste Management Samples* (Compendium) [DOE-3] which includes sampling, physical analysis, organic analysis, inorganic analysis, and radiochemical analysis methods contributed from DOE analytical laboratories. The range of analyses supported by the Compendium methods is included in table 3.3. The Compendium radiochemistry methods represent the closest parallel to standardized methods available to date on high level radioactive waste samples. Proficiencies of the methods are discussed within the methods themselves, however, § 3.3.4 of this report addresses the quality of data that can be achieved with current radioassay methods such as those included in the Compendium. The Compendium is recommended as the resource for which to base analytical method requirements for radioassay of RH-TRU waste. It is also recommended that further compilation of standardized type procedures which are not available in the Compendium be performed before the RH-TRU QAPP is developed.

Table 3.3 Range of Analyses Addressed by DOE Methods Compendium Procedures

Chemical Separations/Analysis		**Instrumental Analysis**
Pu isotopes	^{99}Tc	Gamma Spectrometry
U isotopes	^{210}Pb	Gross Alpha Analysis
Am/Cm isotopes	^{228}Ra	Gross Beta Analysis
^{59}Ni	^{94}Nb	Alpha Spectrometry
^{63}Ni	^{129}I	Beta Liquid Scintillation Analysis
^{89}Sr	^{131}I	
^{90}Sr		

3.3.3 Instrumentation

The counting instruments identified in Appendix D are appropriate for application to RH-TRU waste. Appendix D identifies typical counting instrumentation available in counting rooms across the DOE complex along with a description of the general use of the instrumentation. Counting instrumentation is not biased toward varying levels of radiation pending sufficient counting statistics (which can be controlled by sample geometry and count time). If high count rate is a concern, the sample can simply be diluted or moved to a different geometry. Therefore, the evaluation of applicability of typical counting equipment to the measurement of an RH-TRU waste sample is not necessary.

3.3.4 Data Quality

As was stated in § 3.3.1, the most critical limitation to data quality in radiochemical analysis is the representativeness of the sample. The difficulty in obtaining a representative sample is escalated for

RH-TRU waste based upon the very nature of the waste. Therefore, it is recommended that the risks of requiring a certain level of quality control acceptance criteria be weighted against the benefits. Applying rigorous QC acceptance criteria requirements adds no value to the overall data quality if the sampling method does not parallel the same rigorous criteria. To date, the majority of documented sampling and analysis data on RH-TRU wastes is on the ORNL RH-TRU sludge. Ten years of sampling and analysis data for sludges is documented in ORNL reports [ORNL-11]. Based upon experience and risk benefit, a level of analytical laboratory quality control has been implemented for the analysis of ORNL sludge waste. The level of analytical laboratory quality control which is being obtained to date with the typical counting instrumentation and analytical methods consistent with those provided in the Compendium is described in the following sections and is recommended as the basis for establishing QC acceptance criteria of RH-TRU destructive radioassay.

Instrument Precision and Accuracy

Guidelines for monitoring precision, accuracy, and backgrounds of typical counting instrumentation are provided in the *Standard Methods for the Examination of Water and Wastewater* [APHA-1] and are appropriate in the radioassay of RH-TRU waste. In general, calibration verifications are required for counting instrumentation. Daily calibrations are not needed or necessary for most counting instruments; calibrations are typically performed as needed or recommended in instrument manuals. Therefore, calibration verification standards are used to monitor daily changes in instrumentation for precision determinations and for efficiency checks to determine accuracy.

Method Accuracy and Precision

Method accuracy for RH-TRU samples is measured by analyzing lab control samples (LCS) and matrix spikes for each batch of 20 samples or less. Laboratory control samples typically involve liquid or solid matrices containing the nuclide of interest or mixtures of radionuclides similar to a that expected in a sample. The LCS is applied to the same sample preparation and analysis procedures as a sample. Matrix spikes are prepared by spiking a sample aliquot or diluted sample aliquot with the radionuclide(s) of interest or radionuclide(s) similar in chemistry to that of interest at activity levels within the same order of magnitude as the activity expected in the sample aliquot or sample aliquot dilution. Alternatively, a gravimetric carrier may be used to determine recovery. LCSs and matrix spikes are only required if a chemical separation and/or sample evaporation is performed on the sample, or in some cases when liquid scintillation counting is employed. Measurements which do not require sample preparation (other than dilution) only need to be confirmed with calibration checks of the instrument; one example is gamma spectrometry. Method precision is measured using duplicate samples which may be matrix spike duplicates or sample duplicates, at least one per batch of 20 samples. Attainable quality control limits for method accuracy and precision are included in table 3.4 below.

Table 3.4 Attainable Method QC Limits for Destructive Radioassay

QC TYPE	CRITERIA

Lab control sample	80-120% recovery
Matrix spike	70-130% recovery
Duplicate	≤ 30% deviation

Detection Limits

Method detection limits for radiochemical measurements are dependent on both sample matrix and count time. The accepted standard convention for calculating method detection limits for radiochemical measurements follows the Environmental Measurements Laboratory Procedure [EML-1].

Performance Demonstration Programs

The proficiency of laboratory analysis is typically demonstrated with performance demonstration programs. The WIPP performance demonstration program, implemented for non-destructive assay, is not appropriate for destructive radiochemical analysis. There are other programs currently in place which allow for demonstration of radiochemical analysis proficiency. Those programs include: The Environmental Measurements Laboratory Program (EML) [EML-2] and the DOE Multianalyte Performance Evaluation Program (MAPEP) [DOE-4]. The EML program is primarily used for environmental level radiochemical analysis whereas the MAPEP includes analysis of performance samples which more closely resemble DOE waste samples. Each program provides a report of the laboratory's performance on each individual analyte reported. A laboratory may receive an "acceptable", "warning", or "non-acceptable" ranking for a particular parameter. The EML program evaluates laboratory performance based on historic data distributions from several years. Each laboratory is ranked for each parameter reported based upon it's percentile ranking of a cumulative normalized distribution (i.e. a reported result which is < the 5^{th} percentile and > than the 95^{th} percentile, that is, the outer 10% of the historical data, would receive a "non-acceptable" ranking). In contrast the MAPEP program evaluates laboratory performance based upon the bias of the laboratory's reported result from the reference value determined by the MAPEP program (i.e. a reported result with a bias of >30% from the reference value would receive a "non-acceptable" ranking). While the EML program is widely accepted and provides a quality check of instrumentation, the MAPEP is likely to be more applicable to monitoring the radiochemical methods used for analysis of RH-TRU waste based upon the matrices of the blind controls used in the program.

4.0 RH-TRU WASTE GENERATOR SITE INFORMATION

4.1 Scope and Limitations

The information provided in this section was obtained from DOE sites possessing RH-TRU waste. It was obtained by direct interviews with site personnel, review of process knowledge and the TWBID [CAO-8], and facility tours during site visits. Not all RH-TRU waste generator sites were visited. Due to time and budget constraints, selectivity of site visits was warranted. With the exception of the Hanford Reservation and Battelle Columbus, the primary RH-TRU generator sites were visited. This section is intended to provide a current overview of the status of the RH-TRU generator site radioassay information, fully recognizing that plans, schedules, and funding issues are highly dynamic and thus can become outdated quickly.

Table 4.1 - RH-TRU Generator Site Contacts

Site	Date Visited	Contacts	Phone	Area of Expertise
INEEL	Aug. 26-27, 1996	Mark Dehaan Jack Hartwell Ron Larson Henry Peterson Craig Tyler	(208)-526-2983 (208)-526-9366 (208)-526-0586 (208)-526-8657 (202)-526-1132	TWBID NDA RH engineering ANL-E waste RH-TRU manager
ANL-W	Aug. 27-28, 1996	Steve Aumier Bob Benedict Dave Duncan Carla Dwight Roy Grant Tom Zahn	(208)-533-7479 (208)-533-7166 (208)-533-7487 (208)-533-7651 (208)-533-7400 (208)-533-7217	TD section mgr.-NDA Treatment process TWP project mgr. Waste Programs mgr. TWBID RSWF mgr.
ORNL	Sept. 24-26, 1996	Jeff Gilpin John Keller Tom Monk Sharon Robinson Fred Schultz Jim Stokely	(423) 571-2844 (423) 574-4886 (423) 574-0660 (423) 576-4195 (423) 574-0660 (423) 574-0660	TRU Waste Tracking Destructive assay TRU program mgr. Characterization data NDA Destructive Assay
LANL	Nov. 12, 1996	Ken Coop Bruce Lebrun Toby Ramero Larry Field Steve Betts Nelson Stalnaker	(505)-667-5372 (505)-667-6438 (505)-667-4653 (505)-667-0919 (505)-667-7266 (505)-667-9641	NDA TWBID Packaging TWBID NDA Destructive assay

A complete evaluation of an RH-TRU generator site waste certification and shipment readiness requires more lengthy reviews of the site's RH-TRU radioassay data, radioassay technology, and review of other data relevant to characterization such as gas generation and resource conservation and recovery act (RCRA)

constituents. Table 4.1 is a summary of the sites visited along with the names of the sites' primary RH-TRU waste contacts and phone numbers.

4.2 Introduction to Site's RH-TRU Waste

Section 1.2 pointed out the obvious difference between CH and RH waste: contact dose rate. Section 2.2.1 contrasted CH with RH waste in terms of repository performance. What is discussed in this section is the origin of RH-TRU waste, how it is considered to be more associated with a specific process than the CH-TRU counterpart, what the volume and radionuclide contents are on a site by site basis, and how the radioassay estimates were determined.

Figure 4.1 illustrates the origin of RH-TRU waste, which is typically from hot cell facilities, or liquid-waste processing facilities that service them. These hot cells are used for essentially one of two tasks: separation of isotopes from irradiated targets, or test and evaluation of reactor experimental fuel pins in support of national defense activities. Hot cells are generally used to reduce personnel exposure from highly penetrating radiations emitted by irradiated targets or fuel, hence, the term "remote handled." Examples of these operations are ^{252}Cf separation from Cm-targets at ORNL, and metallurgical testing of EBR-II fuel at ANL-W. Liquid-waste processing systems that have serviced these hot cells contain sludge, which in most cases, is significantly radioactive from the presence of fission and activation products. The relative amount of special nuclear material managed as waste is kept to a minimum because every effort is made to retain or recover the transuranic radionuclides in the "product" stream, rather than in the "waste" stream[6]. Due to this minimization, the specific activity of most transuranic radionuclides is low, RH-streams are dominated in radioactivity by mixed fission and activation products, completely consistent with the baseline inventory report.

The processes that lead to the production of RH-waste are mechanical or chemical, or a combination of the two. The individual processes are well separated conceptually. Mechanical operations include fuel cutting dejacketting (dejacketting may also be a chemical dissolution process), decladding, and embrittlement testing. These operations lead to the presence of experimental fuel residue from chunks, pins, fines, turnings, cuttings, and activated products from metal alloys. In many cases these were fairly dry operations whereby cleanup simply consisted of placing components into small cans and then packaging the small cans in larger storage containers such as 30- and 55- gallon drums. Chemical operations such as acid dissolution, solvent extraction, precipitation, and ion exchange lead to the production of the common liquid waste form, which when made caustic for storage, forms a sludge. Another waste byproduct of chemical operations are the liquid filters, ion exchange resins, and bulk contaminated equipment. It has been common practice to package mechanically-produced wastes as is, with some size-reduction required to meet package-size limitations. Waste produced from chemical processes may be either packaged as is, or treated, as seen fit by the each site.

[6] An exception to this is ORNL where the TRU radionuclides are in relatively small quantity and are not desired in the product of a stream of ^{252}Cf. This is in stark contrast to CH waste, where the TRU isotopes are considered product.

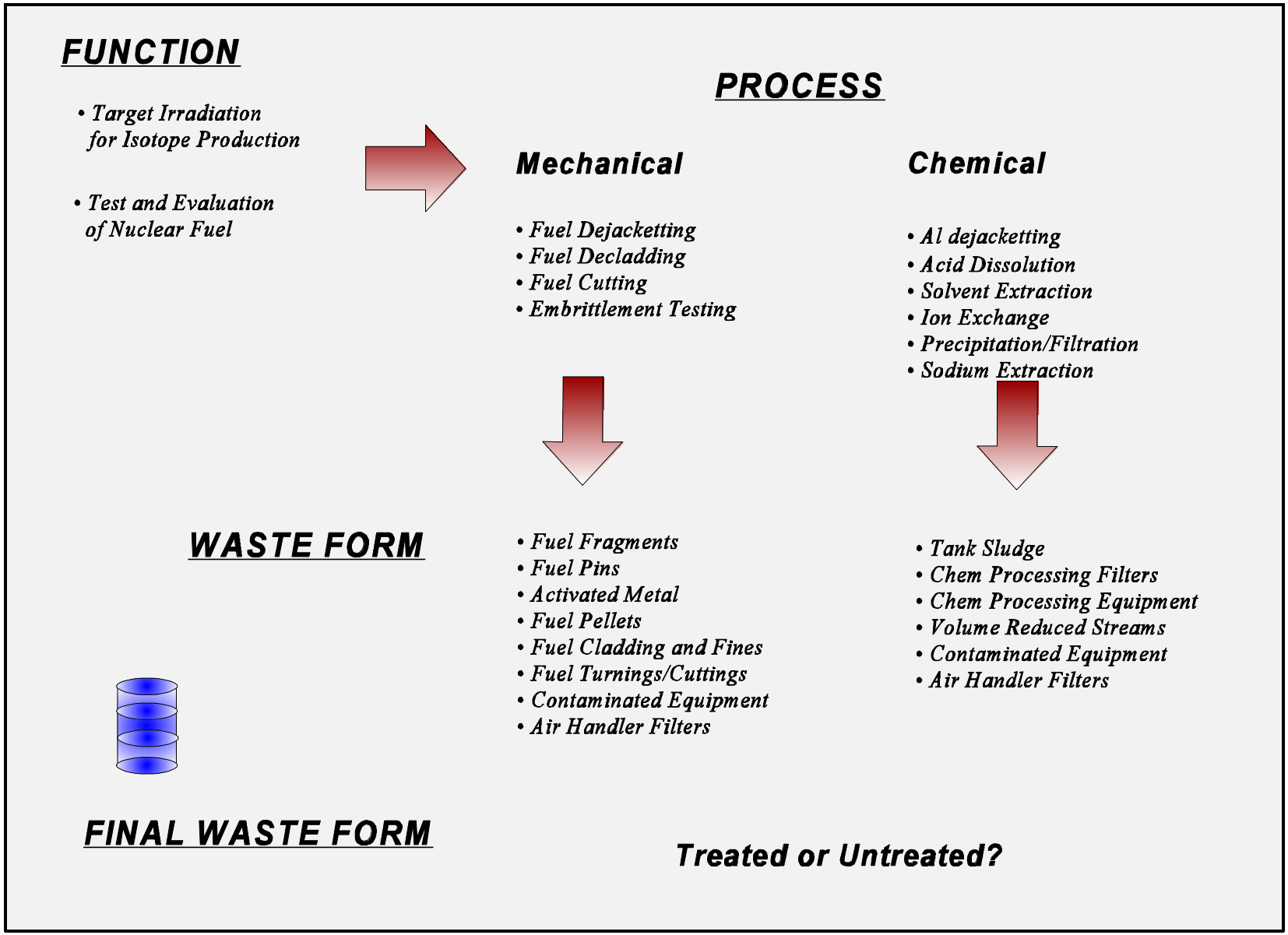


Figure 4.1. Origin of RH-TRU Waste

Because the origin of RH-TRU waste may be compartmentalized into that which results from dry, mechanical processes, or wet, chemical processes, the approaches taken for the collection of radioassay data at an established quality level will be different. Keep in mind that further distinctions between CH and RH will become apparent as the following subsections on inventory are presented, namely:

- Radioisotope proportions for RH waste are very different from CH waste. The predominant radioactivity in CH waste is from plutonium and americium. The predominant radioactivity in RH waste is from mixed fission and activation products (see §2.2.1).

- Except for debris waste generated at ORNL, the neutron emission rate from RH waste is negligible. The gamma-ray emission rate is typically significant with dose rates on small cans in the 100's of rem per hour, on contact;

- Radioisotope dynamics in RH waste are significant relative to CH waste. The predominant radioisotopes in RH are decaying at a faster rate, hence the radioisotopic proportions are rapidly changing; on a relative time scale;

- The use of process knowledge, namely the use of fuel depletion codes and codes for computing activation products in cladding and fission products in fuel is widely used in the management of RH waste;

- RH debris waste is normally packaged in small containers: ORNL uses a 3-gallon polyethylene bucket, ANL-E uses a 7.5-gallon steel container, and LANL uses a 1-gal carbon-steel container; [ORNL-1, INEEL-3, LANL-2] and

- A small fraction of radioactivity in RH sludge is from transuranic radionuclides, as opposed to CH-process streams involving weapons-grade plutonium.

4.3 Site RH-TRU Waste Stream Inventories and Characteristics

The *National Transuranic Waste Management Plan*, [CAO-2] documents the RH-TRU waste storage locations and volumes of both stored and projected RH-TRU waste before treatment and repackaging. Figure 4.2 illustrates the stored RH-TRU volume distributions based upon the values in the plan. As shown, the majority of RH-TRU stored waste, by volume, is located at the Oak Ridge National Laboratory (ORNL). The ORNL RH-TRU waste is basically in the form of two waste types, 58% as inorganic sludge in underground storage tanks and 42% as debris waste in containers. The second largest volume of waste identified is BCL waste, however, this waste may be categorized as commercial fuel and not RH-TRU waste. The remainder of RH-TRU stored waste is primarily debris waste resulting from experimental fuel pin examinations, analytical laboratory operations, and miscellaneous hot cell and glove box operations. Sections 4.3.1 thru 4.3.4 summarize the results from site visits regarding site waste stream types and generating processes.

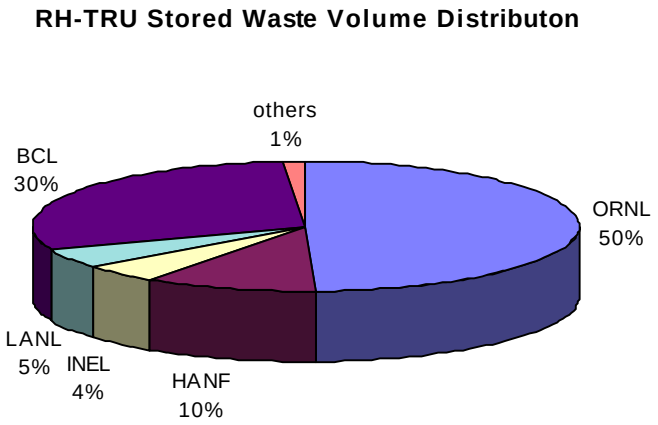


Figure 4.2 RH-TRU Stored Waste Volume Fractions [CAO-2]

4.3.1 Argonne National Laboratory-West (ANL-W)

Several currently stored and future waste streams are identified by ANL-W, however, some are in the process of being re-classified as high level waste**.** The majority of the waste is primarily solidified inorganics and metals of experimental fuel residue waste generated from electrometallurgical treatment R&D and operations. The waste contains high levels of reactive sodium metal. The sodium results from the sodium coolant used in the reactor from which the fuel pins were taken as well as the use of sodium as a heat transfer medium within the fuel pin jackets. Treatment is necessary to remove the sodium. This will most likely result in the waste being defined as newly generated which in turn will affect the designation of waste streams. One current waste stream of approximately $7m^3$ in total volume results from analytical samples, EBR-I waste and subassembly hardware and does not require sodium removal. This waste is currently in storage in steel containers.

4.3.2 Idaho National Engineering and Environmental Laboratory (INEEL)

INEEL RH-TRU waste is comprised of 97% heterogeneous debris from the irradiation and destructive examination of experimental fuel and cladding at Naval Reactors, ANL, and INEEL facilities and 3% homogeneous solids from the flushing of laboratory tanks and process piping at the INEEL's Test Reactor Area. Within these two groups of waste, eleven RH-TRU waste streams have been identified by INEEL, with two of the waste streams encompassing approximately 90% of the INEEL RH-TRU waste by volume. These two waste streams are heterogeneous contaminated debris waste from the Alpha/Gamma Hot Cell Facility at ANL-East. It is packaged in 30- gallon drums, and can contain ^{235}U, ^{239}Pu as metal or ^{239}Pu as oxide. Other Pu isotopes, including ^{240}Pu, are also present, but in small abundances. Fuel remnants including cuttings and filings are not discarded as waste. The waste results from contamination of the material used in clean-up of the destructive examination of fuel pins [INEEL-4].

Four other waste streams also result from destructive examination of fuel but originated from ANL-W hot cell waste and are packaged in cans and 55- gallon drums. Other waste streams stored at INEEL include heavy metal sludge and waste ion exchange resin from the INEEL test reactor area, hot cell debris and laboratory grinding sludge from destructive fuel examination processes at Bettis Laboratory, and sludge waste from the Idaho Chemical Processing Plant (ICPP) from post irradiation examination of H.B. Robinson fuel at the INEEL test reactor area. The ICPP waste is expected to be unacceptable to WIPP WAC with the final definition of "Defense TRU".

4.3.3 Los Alamos National Laboratory (LANL)

The primary RH-TRU waste stream at LANLwas evaluated: hot cell debris waste containing mixed metal scrap and incidental combustibles packaged in ~ 1 gallon sized stainless steel cans. A portion of the cans are packaged within 17 canisters. Other cans (~200) are stored in underground storage shafts. The waste consists of mostly metal or metal equipment, either whole or sectioned and results from destructive examination of experimental fuel pins from the EBR-II breeder reactor. [LANL-1]

4.3.4 ORNL RH-TRU Waste Stream Description

Nine waste streams were identified by ORNL. Four waste streams are sludge waste located throughout four separate sets of tanks, hence four waste stream identifications. The remaining five waste streams are heterogeneous debris waste stored in ~301 concrete casks, 2 drums, and 1 box and are designated as five different waste streams due to unique characteristics of each stream such as presence of RCRA, or liquid, and storage location (i.e. trenches vs. bunkers). The sludge was generated from inorganic solids settled from wastewater in underground tanks from research and radioisotope fabrication, whereas the debris waste results from hot cell, glove box, lab, and maintenance operations from isotope separation and purification. The nature of the debris waste at ORNL is quite different from all other DOE sites [ORNL-1, ORNL-16]. The ORNL RH-TRU waste generation process is largely related to the recovery and purification of transplutonium isotopes from irradiated Cm targets. As a result, radiation intensities (high neutron) and isotopic ratios are significantly different from reactor fuel-type operations.

4.4 Site Radioassay Capabilities

4.4.1 Packaging and Sampling RH-TRU Waste

To appropriately evaluate the capability of a site to sample and package RH-TRU waste, the characteristics of a site's waste stream (i.e. sludge, debris, container type, generating process, etc.) must be considered.

LANL has the capability to package its RH-TRU debris waste. In 1995 LANL completed the assay, packaging, and storing of 17 canisters of RH-TRU waste are currently packaged. However, LANL is not currently set up to sample the 17 canisters or to sample remaining RH-TRU waste cans currently in storage. Capability to sample the 1-gallon sized cans stored in underground sealed shafts could be obtained with some modification of existing hot cell equipment.

ORNL has held sampling and analysis campaigns of its storage tanks containing RH-TRU sludge, but does not possess existing capability to package the sludge. As for ORNL debris waste, the majority of the waste is packaged in non DOT-approved concrete casks. [ORNL-1] There is no current capability at ORNL to sample and repackage existing casks suitable for receipt at WIPP, but there are ongoing activities to develop characterization methods of newly-generated waste by combining DA with NDA results. Special carriers have been fabricated to allow easier handling and transport of the waste. Existing hot cell facilities are used to perform DA analysis. The tasks of packaging and repackaging ORNL's legacy RH-TRU waste is expected to be privatized, as described in § 4.6 of this report.

ANL-W has two hot cell facilities with cell size and shielding capacity for handling RH-TRU 55- or 30- gallon drums of its debris waste and a pneumatic transfer system for delivery of samples to a radioanalytical laboratory. Waste containers that meet existing facility interface parameters could be destructively examined with little effort; some facility modifications would be required to sample 55- and 30- gallon drums of RH-TRU waste in the hot cells. Because sodium removal is required, ANL-W has proposed an upgrade to one of the hot cell facilities which will include the development of a treatment facility to remotely remove sodium before packaging and shipment to WIPP. The ANL-W Remote Treatment Facility (RTF) is currently being reviewed for funding.

The INEEL also has 55- and 30- gallon drums of RH-TRU debris waste, however, the waste is not expected to require treatment. The INEEL has hot cell facilities and equipment with potential capacity for handling their waste drums, and has identified one facility, the Chemical Processing Plant, as an ideal facility for handling RH-TRU waste based upon the location and lack of other mission for this facility. This facility is currently shut down and would need some modification (addition of remote handling and assay equipment) to bring on line for handling RH-TRU waste.

4.4.2 Nondestructive Assay Capabilities

There are no existing capabilities to perform routine NDA measurements on RH-TRU waste. One successful use of NDA on actual RH-TRU waste has been accomplished for measurement of fissile material; all other activities have been research related. In contrast, routine CH-TRU waste NDA capabilities exist at the major DOE facilities----from special nuclear material control and accountability laboratories, to routinely operational facilities for the measurement of waste packaged in 55-gal drums or boxes. Below is a summary of the NDA capabilities throughout the DOE RH-TRU waste generator sites.

a) ANL-W has recently completed measurements of EBR-II assemblies and leached cladding hulls. Passive neutron totals counting measurements were made on nine EBR-II assemblies using a detector subsystem consisting of one ^{3}He detector and one ^{235}U fission chamber. "The measurements indicate that neutron measurements using ^{3}He tubes in a more optimized array (i.e. redesigned instrumented cask or shield ring) could provide useful information with regards to assembly type, total burnup, and plutonium content for EBR-II assemblies. In addition, the

measurements indicate that quantitative information of plutonium content and burnup can be obtained with statistically significant data from an optimized detector system." [ANLW-2] Other passive measurements were made on leached cladding hulls (with anticipated gamma exposure rates in excess of $100Rh^{-1}$) using the *Passive Active Neutron* method. Active measurements have not yet been reported, but the application of this method may be applicable to isotopic discrimination.

b) INEEL has recently performed an evaluation of two NDA techniques for characterization of its primary RH-TRU waste stream using computer models. The first technique was a heavily shielded and strongly collimated HPGe spectrometer system designed using Monte Carlo Neutral Particle (MCNP) modeling. MCNP and synthetic gamma-ray spectrum generators were used to estimate detection limits and precision. The second technique evaluated was the INEEL Passive/Active Neutron (PAN) assay system. For adaptability of the system to RH-TRU waste, a shielded overpack for the drums of the primary INEEL waste stream was designed to shield the detectors from excessive gamma radiation. MCNP modeling was used to estimate detection limits for ^{235}U and ^{239}Pu. Results of the evaluation are reported in [INEEL-5] and address the feasibility of these techniques for RH-TRU waste in which irradiation history and general waste matrices are known. Additionally, INEEL is championing the development of new technologies for the National Spent Nuclear Fuel Program (NSNFP). The proposed technology is termed GNAT/FATS (Gamma Neutron Assay Technique/Fission Assay Timing System). INEEL has recently proposed to apply this technique to RH-TRU waste. [INEEL-6] The technology is based on the detection of fast neutrons and gamma-rays in coincidence, thereby decreasing the effect of high count-rate noise on the signal, and reducing uncertainty from matrix effects.

c) From 1990 through 1994 LANL used a PAN-type system for the measurement of fissile material, primarily ^{239}Pu, packaged in 1-gal sized carbon steel cans. This activity took place in wing 9 of the Ceramic Materials Research facility (CMR). LANL showed that the PAN system, originally developed for contact-handled waste, could be successfully adapted to the measurement of small, 1-gal sized containers, emitting significant gamma-ray dose rates but little neutron dose. The effort is reported in LANL-1 and LANL-2. The prevailing assay issues, such as self shielding, count rate pile-up, and false detection of gamma-rays as neutrons were evaluated. The major design change to the PAN system was the addition of 15 cm of lead in front of the neutron detectors, thus shielding them against the intense gamma-ray field. The RH system was also much smaller (4 ft. x 4 ft. x 3 ft.) than the second-generation PAN system. Other refinements were made, but weren't nearly as crucial. Following the dismantlement of the RH-PAN system in 1994, development efforts of RH-TRU assay systems have ceased.

d) The ORNL has been evaluating the differential dieaway technique for the measurement of RH-TRU under high neutron count-rate conditions, indicative of the RH waste generated by ^{252}Cf production. The feasibility of this method was reported in [ORNL-10] and relies on the use of a neutron source with a factor of 100 higher neutron output than is available in current passive

active neutron systems equipped with D-T neutron generators. The neutron source is a radiofrequency quadrupole (RFQ). The evaluation is still in progress. The ORNL has also demonstrated the ability to use segmented gamma spectroscopy (SGS) to measure gamma-emitting isotopes (predominately ^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{134}Cs, ^{154}Eu, ^{137}Cs) to use with isotopic TRU proportions determined by sampling and analysis activities. The TRU proportion of activity is so small (<2 %) relative to the total activity, that the error in the TRU activity estimate is very large, as reported in ORNL-3 and ORNL-16. In any case, all estimates of TRU concentration are significantly >100nCi/g.

4.4.3 Destructive Assay Capabilities

Analytical capabilities for assay of RH-TRU waste do not require significant equipment upgrade than that required for analysis of CH-TRU waste. The most significant modification involves sampling activities. Site sampling capabilities for RH-TRU waste were addressed in § 4.4.1. Sampling activities for RH-TRU waste must involve coordination with the analytical laboratory to ensure the radiation handling capacity of the laboratory is not exceeded with a sample or batch of samples.

Physical capabilities needed by a laboratory for handling RH-TRU waste samples typically involve hot cells, glove boxes, and radiochemical hoods. Depending on the sample size and radiation level, hot cells and glove boxes may be bypassed, but radiochemical hoods inside laboratories with instituted protective measures for reducing personnel exposure to radiation are mandatory. Sample digestion and preparation of RH-TRU samples need to be performed in hot cells, glove boxes, or radiochemical hoods.

Instrumentation capabilities (at a minimum) for complete radioassay of RH-TRU waste would involve gamma spectrometers, alpha spectrometers, and gas-flow proportional counters or scintillation counters for gross alpha and gross beta measurements. These are typically available in a counting room separated from the sample preparation laboratory.

Laboratories with the above capabilities include ANL-W, INEEL, and ORNL, each having hot cells, glove boxes, radiochemical hoods and complete counting rooms. The LANL analytical laboratory has radiochemical hoods and a complete counting room; however, no hot cells or glove boxes are located within the analytical laboratory. Each of these laboratories are capable of analyzing RH-TRU waste, with special provisions needed for LANL to ensure sample collection and sample digestion are performed within controlled measures prior to analysis in the laboratory. The Radioactive Materials Analytical Laboratory at ORNL and the Chemical Laboratory at INEEL have both demonstrated performance in analyzing CH-TRU waste samples for WIPP during participation in sampling and analysis campaigns of INEEL CH-TRU debris waste and successful completion of the WIPP Performance Demonstration Program [CAO-9] samples for inorganic and organic analysis (radiochemical samples using destructive assay have not been implemented within the WIPP Performance Demonstration Program). The ORNL retains a DOE/CAO approved Quality Assurance Project Plan (QAPjP) [ORNL-15] for laboratory characterization of TRU waste in accordance with WIPP requirements. This project plan was also used as guidance for certain sampling and analysis

campaigns of the ORNL sludges. INEEL has submitted a similar QAPjP to DOE/CAO for review.

4.5 Site Radioassay Data

4.5.1 ANL-W

The ANL-W RH-TRU waste is unique in that the waste requires a sodium removal treatment process which will result in the RH-TRU waste streams being designated as newly generated. This sodium removal process also provides an excellent opportunity for sampling and analysis of the waste before or during packaging of the waste. Due to this opportunity and the absence of ANL-W radioassay data in the TWBID, the evaluation of ANL-W radioassay data is not emphasized in this evaluation. The primary type of data available includes isotopic distribution and estimation of isotopic quantities from ORIGEN codes combined with calculations of the amount of radioactivity expected to remain in the RH-TRU waste after the sodium removal process.

4.5.2 INEEL

Extensive process knowledge is available on the two primary waste streams at INEEL, the alpha-gamma hot cell (AGHC) waste from ANL-E, which makes up approximately 90% of the INEEL RH-TRU waste. Limited process knowledge documentation is available on the remaining waste streams. The significant amount of process knowledge available on the AGHC waste is typical of this type of waste, i.e. residue from the metallurgical examination of irradiated fuel samples. Activities involving the examination of irradiated fuel are typically well tracked and documented to account for losses. The type of AGHC waste process knowledge available at INEEL includes;

- irradiated fuel receipt logs describing the fuel and identifying the enrichment values,
- procedures for the sectioning of the fuel pins,
- AGHC facility operations logs recording dates and fuel examination activities,
- fuel examination reports identifying the percent burn-up and fuel composition,
- waste packaging procedures and operations manuals describing the procedure for packaging waste, identifying radiation limits and calculations for fissile content and TRU content,
- waste packaging logs which track the identity of the fuel elements pertaining to a waste package and the estimates for ^{235}U and Pu content per drum, and
- ORIGEN output indicating the expected fission product yield for a given fuel element.

The importance of this documentation is the ability to estimate fissile content, TRU content, and fission product content per drum of waste. Of course assumptions must be made in order to provide these estimations. Due to efforts to recover the majority of fuel element examination residue, it is estimated that only 1.5% of the fuel pin's inventory should be assumed to be waste. It is also assumed that the work area where the fuel sectioning is performed becomes uniformly and non-preferentially contaminated. After making these assumptions and weighing each drum, estimations of fissile

material, TRU, and fission product content per drum can be made using the burn-up information, fuel enrichment values, ORIGEN calculations, and tracking of waste packages to fuel element identities all obtained from the process knowledge documentation described above.

Example documentation provided for 50 drums of AGHC waste records the ^{235}U and Pu content average per drum as 0.56g and 0.24g, respectively. Using the assumed ^{240}Pu to Pu mass ratio of 11.7% [INEEL-5] and converting the weights to ^{239}Pu fissile gram equivalents, the fge per 30 gal. drum is estimated to be less than 1, which is well below the waste acceptance criteria of 325 fge per cask. Using the ^{235}U and Pu contents listed on the waste packaging logs of the 50 drums, the TRU alpha activity is estimated to average 300 nCi/gm on a drum containing 45 kg of waste (waste weight taken from reference [INEEL-5]) which meets the "greater than 100 nCi/gm" waste acceptance criteria limit. Surface dose rate measurements have also been recorded well below the waste acceptance criteria of 1000 rem/hr per canister; none of the AGHC waste drums have been reported to have surface field dose rates above 30 rem/hr. [INEEL-5]. Thermal Power estimations for a waste drum of AGHC waste are provided in Table 4.2. These are rough estimations using a combination of process knowledge resources provided by INEEL. The table shows that the estimated decay heat of the AGHC waste is well below the waste acceptance criteria limit of 300 watts/canister.

On the basis of the review, the logic used by INEEL to estimate TRU and fissile content are technically sound, particularly when applied to a population of like drums. The question is how variable is the characterization data over the entire set of drums--- this is what needs to be investigated and validated by some characterization approach. The process knowledge appears thorough and well documented. However, no assay data is available on INEEL RH-TRU waste to confirm the process knowledge documentation and assumptions (Modeling projects are currently implemented at INEEL to determine feasibility of using NDA for RH-TRU radioassay) [INEEL-5]. Process knowledge and data on other waste streams are limited and will require collection of additional records and/or assay.

Table 4.2 AGHC Decay Heat Estimations (INEEL Data)

Nuclide	Activity* (Ci/drum)	"Q value"** (W/Ci)	heat contribution (W/drum)
Mn-54	3.72E-05	4.98E-03	1.85E-07
Co-60	1.91E-04	1.51E-02	2.88E-06
Sr-90	6.11E+00	1.16E-03	7.09E-03
Y-90	6.11E+00	5.54E-03	3.38E-02
Ru-106	6.69E-04	5.95E-04	3.98E-07
Rh-106	6.69E-04	1.89E-02	1.26E-05
Sb-125	2.15E-02	3.37E-03	7.25E-05
Cs-134	3.35E-03	1.02E-02	3.41E-05
Cs-137	6.11E+00	1.01E-03	6.17E-03
Ce-144	2.96E-04	6.58E-04	1.95E-07
Pr-144	2.96E-04	7.39E-03	2.19E-06
Eu-152	3.12E-05	7.65E-03	2.39E-07
Eu-154	5.61E-03	9.08E-03	5.09E-05
Eu-155	5.24E-02	7.59E-04	3.98E-05
U-235	1.20E-06	2.71E-02	3.26E-08
Pu-238	5.90E-04	3.26E-02	1.92E-05
Pu-239	1.30E-02	3.02E-02	3.93E-04
Pu-240	6.80E-03	3.06E-02	2.08E-04
Pu-241	2.06E-01	3.20E-05	6.59E-06
Total			**4.80E-02**

***Note**

Activity estimates for nuclides in italics taken from reference [INEEL-5] which presumes the waste to be 15 year-aged EBR-II fuel and uses ORIGEN inventory calculations.

Activity estimates for Rh-106 and Pr-144 are assumed to equal that of their parents, Ru-106 and Ce-144, respectively.

Activity estimates for Sr-90/Y-90 are assumed to roughly equal that of Cs-137.

Activity estimates for U-235 and Pu-239-241 taken from INEEL waste operations for 50 drums; values are not decay corrected.

Activity estimate for Pu-238 assumes the activity ratio of Pu-238 to Pu-239/240 is equal to roughly 0.03 based upon ORNL assay of experimental fuel from Rocky Flats; values are not decay corrected.

**Q values (Watts/Curie) taken from Table B.1, Appendix B, of the Department of Energy Office of Environmental Management Integrated Data Base Report, document DOE/RW-0006, Sept. 1995. (Web site: http://www.em.doe.gov/idb95/tabb1.html, updated 3/13/96)

4.5.3 LANL

LANL radioassay data for thirteen canisters of packaged RH-TRU waste is well documented. Data on the other 4 canisters (of 17 total) was not provided. As mentioned earlier, the primary tool used for the measurement of ^{239}Pu and ^{240}Pu was a passive active neutron device, tailored specifically for the measurement of 1-gal cans, emitting high gamma-ray dose rates but small neutron dose rates. A complete description of the system, how it was tested, and how it was calibrated is presented in LANL-1 and LANL-2. The system was built and operated by experts in this technique; they calibrated the system to account for self-shielding and for gamma-ray discrimination. The active mass was reported on the largest majority of the cans with mass **less than** 1 gram. The passive result was used for reporting the mass when self-shielding was thought to be significant, i.e. in cases when the mass was many tens of grams of ^{239}Pu.

grams of ^{239}Pu. During the project, each can was measured and then staged in drums, prior to placement in the RH-canister. By standard accounting procedure, the cans were placed in each canister such that the sum of the measurement and its error was less than the 325-g canister limit.

Results of the 1-gal can-size measurements were recorded in tabular form as the fissile assay mass and associated error. Results for 376 1-gal cans were provided. For the 13 canisters the following results apply for fissile mass, as seen in figure 4.3: mean = 204 g, avg. std. dev. = 46 g, avg. rel. error = 14%

These results were consolidated from data given by Coop and Romero as follows[7]. Each 1-gal can was measured and reported with fissile mass, m_i, and the associated error, at one standard deviation for the measurement reported as s_i. For a canister, the total fissile mass of each can is summed and the error is computed in a root mean square fashion:

$$Mass\ of\ fissile\ mass\ in\ canister = \sum_{i=1}^{n} m_i$$

$$with\ corresponding\ error\ (at\ 2\sigma) = 2\left(\sum_{i=1}^{n} s_i^2\right)^{\frac{1}{2}}$$

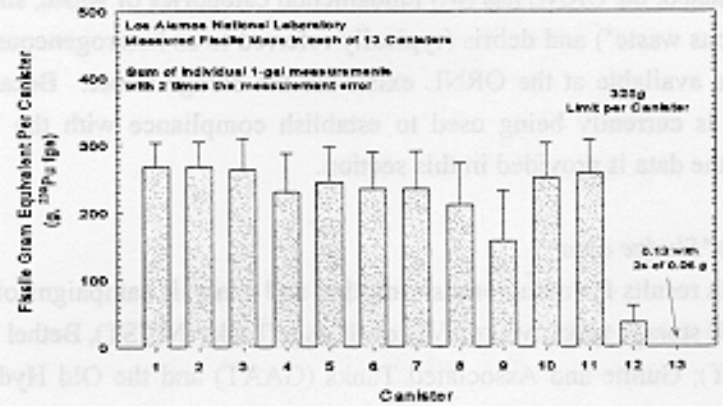


Figure 4.3. Reported Nondestructive Assay Results of Fissile Gram Equivalent in 13 Stored Canisters

[7] T. Romero provided an EXCEL spreadsheet of the 1-gal can data (376 cans). Dr. K. Coop provided system data sheets for each of 364 1-gal cans as well as all internal memos related to the measurements. D. Christensen provided search results of RH-TRU waste from the LANL waste certification database.

As seen in figure 4.3, the mean fissile mass plus two times its error is less than the 325 fge limit. In all cases the results were reported as being less than the most essential waste acceptance criteria, a fissile gram equivalent of 325 grams per canister.

Other records were kept. Fission product inventory was scaled to the PAN measurement; review of data records showed that the fission-product fractions were constant across all thirteen canisters, and included the isotopes of ^{239}Pu, ^{235}U, ^{137}Cs, ^{90}Sr, ^{90}Y, ^{106}Ru, ^{106}Rh, ^{125}Sb, ^{125m}Te, ^{137m}Ba, ^{147m}Pm, and ^{155}Eu. These fractions were determined initially by sampling the fuel fragments, and then using the measured ratios for the duration of the project. The assay results show that all isotopic ratios are thereby constant. Additional data kept were real-time radiography examination of each 1-gallon can.[8]

A cursory technical review of the methods and the data was performed. The measurements were performed nominally. Accounting records were relatively well managed; a detailed review probably needs to be performed that ensures the raw data was transcribed properly into the several systems that combined and tracked the data. No glaring errors were noticed, but it was difficult to account for the lineage of each and every 1-gal can. The measurements were technically sound and adequately described. At this stage of the process, there would be zero added value in performing any repeated measurements of the 13 canisters for which data exists. A data validation effort would complete the process of determining the extent (i.e. degree of quality) to which the LANL waste meets all WIPP WAC.

4.5.4 ORNL

As previously stated, the ORNL has two fundamental categories of waste, sludge (typically referred to as "homogeneous waste") and debris (typically referred to as heterogeneous waste). The majority of radioassay data available at the ORNL exists for the sludge waste. Because the sludge data is so extensive and is currently being used to establish compliance with the WIPP WAC, a thorough description of the data is provided in this section.

"Homogeneous" Sludge data

The sludge data results from numerous sampling and analysis campaigns of the four different sets of ORNL RH-TRU storage tanks; Melton Valley Storage Tanks (MVST), Bethel Valley Evaporator Service Tanks (BVEST), Gunite and Associated Tanks (GAAT) and the Old Hydrofracture Facility Tanks (OHFT). Both the supernatants (not considered RH-TRU) and the sludges within the tanks have been sampled and analyzed. The data collected on the sludges from campaigns occurring between 1985 and the early part of 1996 are summarized in an ORNL technical report, *Statistical Analysis of ORNL LLLW System Transuranic Wastes* [ORNL11]. Various other reports are also available on individual campaigns between 1985 and early 1996, but all sludge sampling and analysis data from these reports are summarized in the ORNL11 report. More recent data collected in the fall of 1996 on the MVSTs is summarized in another ORNL technical report, *Characterization of the MVST Waste Tanks Located at*

[8] Private communication with L. Field, LANL

ORNL [ORNL12]. New data has recently been collected for two other sets of tanks, the BVESTs and the OHFTs, for which similar reports have been published in early 1997 [ORNL 13 and ORNL 14]. For this evaluation, however, only reports ORNL11 and ORNL12 have been used.

The data included in ORNL11 represents statistical analysis of the best data possible from that obtained from the various sampling and analysis campaigns up until Fall 1996. The report states that the summarized data was not taken as part of a comprehensive characterization of the ORNL waste tanks, but the data represents many different projects with different needs, analytical requirements, and data quality objectives. In general, the more recent the data, the more accurate the data due to improved laboratory technology and better familiarity with the samples. The data collected represents a wide variety of analyses. Analyses were performed at the Radioactive Materials Analysis Laboratory and include physical measurements, chemical measurements, radiological measurements and measurement of organic compounds.

U.S. Environmental Protection Agency (EPA) methods[9] were used by the analytical laboratory to perform sample digestions and gamma spectrometry measurements. Gross alpha, gross beta and radionuclide measurements were performed using in-house methods due to the lack of available EPA standardized methods or due to the inappropriateness of the standardized methods to high level radioactive samples. The level of quality assurance applicable to the data identified in the ORNL11 report varies depending on the time period of the data. The most recent data identified in the ORNL11 report, data collected during early 1996, corresponds to a level of analytical laboratory quality assurance which parallels the proposed level of quality assurance described in § 3.3 of this report. According to ORNL staff, the data included in ORNL11 was used to generate the radionuclide quantities listed in the latest revision of the TWBIR. [CAO-8]

The ORNL12 report includes data from the fall 1996 sampling and analysis campaign of the MVSTs. The purpose of this campaign was to obtain additional data needed to address waste processing options, further evaluate the waste characteristics with respect to the WIPP WAC and Nevada Test Site WAC (NTS is the possible destination for tank supernatants), address criticality concerns, and to further evaluate the ability to meet transportation requirements. The significance of the ORNL12 report is not only the inclusion of more comprehensive data, but it uses the data to establish upper boundary limits for fissile gram equivalent activity, plutonium-239 equivalent activity, and thermal power generation. The estimates are based upon packaging wet sludge in 55-gallon drums and assume the samples from which analyses were made are representative of the tank sludge contents. Because the packaging process is unknown at this time and may involve dilution or concentration of the sludge by drying, these estimates are only preliminary. Estimated values are included in Appendix E and are estimated per waste tank. The estimates were found to be far below the WIPP WAC limits currently proposed for RH-TRU waste.

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[9] Sample digestion and physical measurements include those from the EPA guidance manual, *Test Methods for Evaluating Solid Waste* (SW-846). EPA Method 600/901.1 was used for gamma emitting radionuclide measurements.

The statistical analysis data in ORNL11 indicates that the beta activity in the ORNL sludge is dominated by Cs-137, Sr-90, and Eu-152. Eu-152 currently contributes ~20% of the total beta activity in the ORNL sludge, however it's significance to the WIPP PA is negligible due to its short half-life of 13 years. Dominant alpha emitters within the sludge include Cm-244, Cm-243, Am-241, Pu-238, Pu-239, and U-233[10]. Like Eu-152, Cm-243 and Cm-244 are short lived, with half-lives of 29 years and 18 years, respectively. This fact makes these isotopes less significant to the impact on the WIPP PA. With these factors considered, the primary radionuclides in ORNL sludge impacting the WIPP PA are consistent with the list of radionuclides listed in the WIPP PA with highest importance rankings (refer to § 2.2.1 of this report).

In this evaluation, the radiological data reviewed was found to be technically reasonable based upon the methodology for performing analyses. The range of data collected also represents the amount of radioassay data needed in order to calculate WIPP WAC for nuclear properties (excluding dose rates, which should be measured on the final package versus calculated). The data also includes individual radionuclide quantities, which make up at least 99% of the alpha activity and 99% of the beta activity and include the measurements needed to determine the waste to be transuranic. This amount of information provides the data necessary for inclusion in the TWBID for which the WIPP performance assessment is calculated. The critical limitation of this data includes the sampling statistics. Typically, the major data limitation with sampling and analysis of waste is the representativeness of the sample. The ORNL11 report indicated that for the MVSTs, BVESTs and OHFTs, the sludge has only been sampled in single locations within the tanks due to limited access. Many of the GAAT tanks had sludge samples taken at three different locations with large differences in concentration observed from most species measured. It has also been noted in the ORNL11 and ORNL12 reports that individual sludge cores from the tanks showed different layers, indicating the sludge is not homogeneous. Therefore, the data is biased. The data is also subject to change due to potential waste packaging/treatment processes. However, this change may be calculated with waste treatment process control.

Debris Waste Data

Measurements of debris RH-TRU waste from the Cm-target processing facility at ORNL, the Radiochemical and Engineering Development Center (REDC), have been made for several years. Since the mid 1980s REDC has reported isotopes according to a nominal test to relate isotopic concentration with neutron and gamma-ray dose rate; hence, most of the activity reported is ^{244}Cm., with little emphasis on the TRU radioisotopes. All of the TWBIR data for ORNL debris waste was submitted using this methodology, although the authors are aware of some limited NDA data on file for CH-TRU waste produced from this process over the years. In FY96, a significant effort was initiated [ORNL-2] to smear and sample debris waste, and then perform rigorous destructive assay on each smear. Radiochemistry measurements of contaminated-surface smears collected from within the REDC process and waste storage hot cells were statistically analyzed to estimate the proportion of gross alpha activity contributed by the

[10] Dominant radionuclides are based upon those that contribute at least 1% towards the total alpha or total beta activity value calculated in the "Weighted Statistics Over All Tanks" table in reference ORNL1.

transuranic radioisotopes ^{246}Cm, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am, and ^{243}Am. Unlike weapons-grade plutonium, waste generated from the REDC is composed largely of non-TRU, alpha-emitting radioisotopes ^{244}Cm and ^{252}Cf; hence the classification as TRU-waste by alpha-activity concentration alone, is difficult and requires a thorough evaluation of radiochemical results using statistical methods that address the unique aspects of the problem. This work was performed to measure and report isotopic ratios of TRU radionuclides and ^{252}Cf to gross alpha activity; test whether the ratios are constant over different types of debris waste; assist in the calibration of nondestructive assay instruments; and establish a basis for ascertaining whether there is any evidence to suggest that the concentration of TRU radionuclides is less than the 100 nCi/g limit for classification as non-TRU waste.

The smear project was started in January of 1996 with a collection of Q-tip smears from 9 hot cells at the REDC. The project plan [ORNL-2], describes how the debris waste was sampled and analyzed. A total of 60 smears were taken from 5 separate classes of waste: glass, metal, polyethylene, cell surface, and wipes to determine whether a particular ratio was dependent on material type. Gross alpha analysis, alpha spectrometry, gamma spectrometry, and gross neutron counting were performed on the smears. Two sets of duplicates were also analyzed for evaluating variability in sample collection. Additionally, a sample was leached. The leachate was analyzed to allow for the intercomparison of isotopic raios between health physics smears (contamination collected from a fraction of the entire surface of an object) and complete sample analysis (contamination for the surface and volume of entire object).

The results indicated that the ratio of ^{244}Cm alpha activity to gross alpha activity was greater than 0.90 in 93% of the observations. Additionally, the ratio of all TRU-radionuclide activity to gross alpha activity was found to be 0.0227 or 2.27%. The results are within the documented process knowledge: the contribution of alpha activity from TRU radionuclides is very small, with the overwhelming majority of alpha radioactivity from ^{244}Cm. As one would expect, for the smears collected, the proportion of alpha activity that is TRU was not statistically different for differing materials. A linear regression of the proportion TRU data showed that the proportion TRU decreases with increasing alpha activity, but only slightly.

The second phase of this project was reported in ORNL-16. 3-gal buckets of waste were analyzed by segmented gamma-ray scanning. The easy-to-measure isotopes of ^{106}Ru, ^{110m}Ag, ^{125}Sb, ^{134}Cs, ^{154}Eu, and ^{137}Cs were accurately quantified by SGS. The SGS system was retrofitted to allow for the accurate analysis of waste reading 100 mrem/h (roughly 50 gamma and 50 neutron). A separate statistical analysis of the radioisotopic fractions was performed using both parametric and nonparametric techniques. Ratios of TRU isotopes to the six “easy-to-measure” were developed and computed as geometric means and standard deviations (lognormal distribution) and as percentiles (nonparametric analysis). Additionally, one of the 3-gal buckets was ashed. The ash was analyzed by radiochemical means. The radiochemical results of the ash indicate a TRU activity concentration of 500 nCi/g. All estimates by NDA, when combined with the statistical analysis of isotopic ratios were above 160 nCi/g and below 2000 nCi/g. There is no evidence to suggest the waste is not TRU. It is anticipated that this work will continue at some

nominal level of support. More data will improve the statistical accuracy of projections made with just a few data points.

4.6 Site Waste Certification/Shipment Plans

This section describes each site's plans for certification and subsequent shipment of their waste to WIPP as they were conveyed during the site visits or contacts. Shipment schedules are documented in the *National TRU Waste Management Plan*. [CAO-2] All plans are pending the development of data quality objectives and subsequent implementation for the characterization of RH-TRU waste.

4.6.1 ANL-W

As stated in previous sections of this report, ANL-W RH-TRU waste contains reactive sodium metal which must be removed before the waste is accepted by the WIPP WAC. ANL-W has proposed the development of the Remote Treatment Facility (RTF) which requires an upgrade to the current Hot Fuel Examination Facility (HFEF). The RTF will be used to characterize, segregate, treat, and/or repackage RH materials, as necessary. Development of the RTF is dependent upon funding approval. The ~7 m^3 of RH-TRU waste which does not require sodium removal will require completed characterization and packaging once the data quality objectives for RH-TRU are established. Schedules for processing of the ANL-W high sodium RH-TRU waste or packaging/characterization of the non-sodium RH-TRU waste were not conveyed during the site visit.

4.6.2 INEEL

Plans currently exist at the INEEL to provide direct certification of RH-TRU waste for WIPP disposal without pretreatment. These plans are based upon two project studies at the INEEL. One project involves the demonstration of using conventional non-destructive assay instrumentation and acceptable knowledge to characterize heterogeneous hot cell debris waste via simulated response modeling systems. The second project involves the demonstration of a new non-destructive assay technique which the INEEL indicates will eliminate the need for extensive prior knowledge of the waste matrix before an accurate NDA can be accomplished. These projects are currently in process. It is also expected that the awarding of a new waste treatment facility, the Advance Mixed Waste Treatment Facility (AMWTF), may accommodate the handling of all or some of the INEEL's RH-TRU waste inventory. This contract is to be awarded soon.[11]

4.6.3 LANL

Los Alamos intends to utilize the robust amount of process knowledge available from fuel-burnup codes in conjunction with the NDA measurements made [LANL-1, LANL-2] of individual 1-gal cans that reside in RH-TRU canisters. These canisters comprise the most likely batch of already assayed RH-TRU waste in the complex and should be ready to ship according to the proposed CAO schedule, provided the recommendations (or needs) identified in § 5 are carried forward. LANL also has a considerable amount

[11] Information on the INEEL RH-TRU certification/shipment plans obtained from *"Status of RH-TRu Activities at the INEEL"* presentation by Craig Tyler, TRU Waste Department, Lockheed Martin Idaho Technologies Company, November 14, 1996, RH-TRU Working Meeting, Albuquerque, NM.

of RH waste stored below grade. The decision to retrieve this material under the purview of a significant ER project has not been made at this time, but it appears likely that the material will stay where it is.

4.6.4 ORNL

The ORNL plans to treat, package, characterize, and ship its legacy RH-TRU and debris sludge waste, liquid low level supernate, and CH-TRU debris waste through the awarding of a DOE privatization contract. Requests for proposals to the contract have been initiated and the awarding of the contract is scheduled for early 1998. The private company being awarded the contract will be responsible for determining the treatment, characterization, and packaging methodology. It is expected that the sludge data will provide the acceptable knowledge needed for characterization with statistical sampling and analysis or non-destructive assay of the final waste form to confirm the acceptable knowledge. The characterization plans for the legacy debris waste are uncertain at this point.

5.0 CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

As was stated in §1.0, the purpose of this report is to provide the information necessary to assist the DOE CAO in development of the data quality objectives for radioassay of RH-TRU waste. The report meets the intended purpose by applying the waste characterization DQO process planning flow diagram by Grumbly in figure 1.1 of §1.0. Table 5.1 identifies the steps of the DQO process planning flow diagram, the sections of the report which apply to those steps, and the important points related to each step.

5.2 Recommendations

Having stated the important points in table 5.1, the following recommendations are necessary to establish DQOs for RH-TRU waste.

1) **The DQOs should require fissile gram equivalent, and possibly thermal power, to be measured directly or calculated based upon analytical measurements. Corresponding errors must be reported.** RH-TRU generators should be required to demonstrate that measurement error has been evaluated to a sufficient quality so that when the error is added to the measured parameter, the result is less than a corresponding WAC.

2) **A DQO should be considered which requires the sites to demonstrate the ability to determine radionuclide ratios within an acceptable error range.**
This recommendation for DQOs is to ensure the WIPP PA calculations are based upon appropriate radionuclide distributions reported by the sites in their TWBIR. This DQO could simply be the requirement to participate in a blind sample program and show demonstration to determine radionuclide ratios within a certain error. An acceptable error range needs to be determined as part of the continuing DQO process. A logical requirement might be that the isotopic ratio error not exceed the error introduced by the performance assessment, for that particular isotope.

3) **Demonstration by the sites that DQO quality control specifications are met for RH-TRU radioassay should rely much more on expert review and site audits, than currently recommended for the case of CH waste.**

4) **Establishing the radioassay requirements and subsequent quality assurance objectives requires further work, listed in order of importance.**

Recommended project 1:
The data packages for 13 of the 17 canisters of LANL RH-TRU waste contain information necessary for satisfying the WIPP WAC. The data for all 17 canisters needs to be validated by expert panel review to ensure that the WAC have been met. This validation must include a review of the NDA methodology, calibration, quality control, and the use of ORIGEN calculations to scale fission and activation product

inventory (along with the initial sampling and analysis scheme). It must also include an audit of the data collection and management process to show traceability of the data packages to raw data measurements. The authors believe that by performing a thorough validation of radioassay, the risk benefits associated with pulling the canisters out of storage, sampling the packaged waste, and remeasuring will not be required. It is likely that the value of a slight improvement in radioassay quality achieved by a risky reanalysis of the waste is so small as to be negligible in satisfying the WAC.

Recommended project 2: (NDA proficiency)

The successful utilization of NDA equipment for RH-TRU assay will require some development effort. The magnitude of the effort will depend on how accurate results have to be. Once NDA systems can be adapted to the more demanding environment of RH-waste, they will be able to ensure that individual container WAC limits are not exceeded, provided good sampling and analysis data is used in conjunction with bulk measurements. As the goodness of sampling and analysis (or PK) data is diminished, NDA systems are forced to work harder and harder beyond reasonable capability. If this is the case, significant development efforts will be required. Thus the need to develop a strategic balance between the tools of radioassay become particularly more focused for RH waste.

Without specifying how these needs are accomplished, the important considerations that need to be worked out for RH-TRU NDA are listed in order of priority:

1. Equipment needs to be adaptable to use in or near a hot-cell facility, in areas with potentially high background irradiation levels. Equipment needs to be simple to maintain and reliable for adaptability to harsh environments.
2. Equipment needs to respond quickly to high count rates. Pulse-pile up and dead-time correction are very important.
3. Discrimination between the signal desired and background noise (or interference) needs to be evaluated in high gamma-ray fields and in high neutron fields.
4. When fuel residues are suspected in the waste, corrections need to be made for self shielding, a potentially significant effect.
5. For neutron based systems, in addition to pulse pile-up corrections, good gamma-ray discrimination is needed. Coincidence counting may need to occur in a much faster time window. Differential dieaway will need a stronger neutron source.
6. For gamma-ray spectrometry-based systems, improvements need to be made to reduce count rate and the effects of Compton scatter, which reduce the sensitivity and resolution of the system. Techniques for measuring the gamma-rays from fission and activation products and relating these radionuclides to TRU radionuclide content need to be validated and improved.
7. Matrix corrections for RH-waste may not be important when properly accounted for in the initial calibration stage and when the waste management plan imposes the following conditions on packaging: for debris waste, measure small containers; for processed-waste, the matrix from drum to drum is constant. If these conditions cannot be met, a significant development effort must be initiated to account for matrix and source distribution effects.

8. Advanced NDA techniques that do not employ the conventional methods, in particular, need to be evaluated carefully prior to funding to ensure that condition 1 is met and that it offers specific advantages in separating signal from background .

9. NDA system design and utilization should be focused on ensuring a measurement can be performed to assure WAC parameters are not exceeded.

Recommended project 3: (Development of a Characterization Strategy)

Obtaining a representative sample of debris waste is impossible without shredding and homogenizing the waste. Representative sampling of non-debris waste (e.g. ORNL sludge), although more possible than debris, has not been demonstrated to date. Therefore, characterization of waste must be carefully strategized to optimize use of available resources. Available resources include process knowledge, nondestructive assay, and destructive assay. A strategy which makes use of all three incorporating quality assurance objectives of all three, needs to be developed for characterization of RH-TRU waste. Developing the strategy will first require a risk assessment of RH-TRU waste (in terms of both debris and non-debris). The second part of the strategy development wi3ll include identifying decisions on the use of process knowledge to specify analysis needs. The third part of the strategy will be to define sampling requirements in terms of debris and non-debris with risk benefit in mind. For non-debris waste, this definition will include the extent of samples to collect. For debris waste this will include a definition of when to obtain samples (via screening techniques and elimination of needs from process knowledge) and how to obtain samples (if at all) to ensure the WAC limits <u>are not exceeded</u>. The strategy for analysis of debris waste will then include only non-destructive assay or destructive assay or a combination of both. The results of the project will be a flow diagram for characterization guidelines in terms of debris waste and non-debris waste with tables identifying quality assurance objectives.

Recommended project 4: (Validate use of calorimetry)

The WIPP Waste Acceptance Criteria (WAC) requires determination of heat generation. When sufficient acceptable knowledge data is not available for a particular waste canister, its thermal power may be assayed directly. Currently, heat generation is computed by summation of individual radioisotope contributions. Typically, this involves a combination of nondestructive assay (bulk measurement of the canister) and destructive assay (sampling and analysis) to obtain measurements of all contributing radioisotopes. However, the process of measuring each isotope individually is expensive, complex, and increases personnel exposure to radiation. This recommended project is to provide the information to validate the use of a direct bulk measurement of heat generation using calorimetry. Direct measurement of heat generation rate affords a definitive level of assurance that the thermal power limit is not exceeded, particularly as the thermal power of a waste stream approaches the WAC limit. Additionally, this direct measurement may provide the most accurate, least expensive, and safest determination of heat generation in RH-TRU waste.

Table 5.1 DQO Process Planning Matrix for RH-TRU Radioassay

DQO Planning Process for Radioassay	Section	Important Points
Identify Issues, Regulatory Drivers	2.0	• DQOs currently established for CH-TRU are inappropriate for RH-TRU. • Emphasis for establishing RH-TRU DQOs should be based more on transportation requirements than WIPP PA requirements. • Neither the transportation requirements nor the PA radionuclide source term specifications require the direct measurement of each and every radionuclide. • The influence of RH waste on the PA (by volume, TRU activity, and release fraction) is a small, nearly negligible fraction of the CH influence. • The most important measurements for RH-TRU will be driven by two transportation requirements: fissile gram equivalent and heat generation. Of secondary importance for the PA is that the relative isotopic ratios be reasonably estimated, but not necessarily measured. • Fissile gram equivalent will be the prevailing DQO, in contrast to the existing DQO for CH-TRU waste, total alpha activity, driven by radiolytic gas generation. • DQO QC requirements should be in place to ensure that transportation acceptance parameters are not exceeded. Radioassay error must be known, reported, and utilized in the certification process, but is not tantamount to making compliant shipments. The closer to a given limit a generator wants to ship, the better the radioassay data needs to be qualified.
Formulate Questions, Possible Answers, Data Needs	3.0	• Radioassay data collection for RH-TRU will require a strategic balance between process knowledge, nondestructive assay, and destructive assay. • Proficiency of non-destructive assay on real RH-TRU debris waste has been demonstrated in one case, LANL. Proficiency for other RH waste streams has not been demonstrated and is not well known. • Proficiency of destructive assay on RH-TRU waste is well known; there is no difference from CH waste. • The primary limitation of RH-TRU waste characterization by DA is sampling, therefore indiscriminate application of rigorous quality control acceptance criteria to analytical measurements adds no value. • It is unlikely that NDA measurements of RH waste will be as accurate os for CH waste, all things being equal.
Compile Existing Data (PK and S&A)	4.0	• RH-TRU inventory can be separated into two basic waste streams: debris and non-debris. • The primary data available for RH-TRU non-debris (sludge) come from sampling and analysis. • The primary data available for RH-TRU debris come from fuel burn-up/ORIGEN calculations combined with dose rate measurements, or passive active neutron measurements (LANL).
Evaluate Existing Data for Sufficiency	4.0	• LANL debris radioassay on 13 packaged canisters is likely to be sufficient for shipping and disposition to WIPP, pending a validation of the data. • ORNL sludge data provides the amount of information necessary to demonstrate WAC, but requires confirmation with random sampling and analysis of final waste form. • INEEL, LANL unpackaged, and ORNL debris waste information sufficiency is dependent upon development of NDA application to RH-TRU waste and a program for the balanced use of PK, NDA, and DA. • ANL-W debris waste will be treated thus allowing several options for characterization that will likely be much easier than for debris alone.

6.0 REFERENCES

[APHA-1]
"Standard Methods for the Examination of Water and Wastewater", American Public Health Association, Washington, DC, current edition.

[ANLW-1]
Mosby W.R., "NDA Laboratory Functions and Capabilities," informal internal report, Argonne National Laboratory-West, 1994.

[ANLW-2]
Klann, R.T., ANL-W internal correspondence, "Preliminary Report: Non-destructive Neutron Measurements of EBR-II Assemblies in FCF", June 16, 1997.

[ANLW-3]
Aumeier, S.E., Poenitz, W.P., and Forsmann, J.H, "Nondestructive Assay of Leached Cladding Hulls at ANL-W", Paper presented at June 1997 American Nuclear Society Conference, Orlando, FL.

[ANSI-1]
ANSI N15.20 Guide to Calibrating Nondestructive Assay Systems

[ANSI-2]
ANSI N15.36-1983, "American National Standard for Nuclear Materials - Nondestructive Assay Measurement Control and Assurance", 1983.

[ASTM-1]
ASTM C1030 Determination of Plutonium Isotopic Composition by Gamma-Ray Spectrometry (Intercomparison in P&B)

[ASTM-2]
ASTM C1128 Guide for the Preparation of Working Reference Materials for use in the Analysis of Nuclear Fuel Cycle Materials

[ASTM-3]
ASTM C1133 Standard Test Method for NDA of SNM in Low Density Scrap and Waste by Segmented Passive Gamma-Ray Scanning

[ASTM-4]
ASTM C1207 Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting

[ASTM-5]
ASTM C1316 Standard Test Method for the Non-Destructive Assay of Nuclear Material in Scrap and Waste by Passive-Active Neutron Counting using a ^{252}Cf Shuffler

[CAO-1]
Brown M.R. "Proceedings of the RH TRU Waste Working Group Meeting, November 14, 1996," United States Department of Energy Memorandum, Carlsbad Area Office, CAO:NTP"MRB 96-2276, December 5, 1996.

[CAO-2]
"The National Transuranic Waste Management Plan," United States Department of Energy Carlsbad Area Office, DOE/NTP-96-1204, rev. 0, September 30 1996.

[CAO-3]
"Remote-Handled Transuranic Waste Study," United States Department of Energy Carlsbad Area Office, DOE/CAO 95-1095, October 1995.

[CAO-4]
"Waste Acceptance Criteria for the Waste Isolation Pilot Plant," United States Department of Energy Carlsbad Area Office, DOE/WIPP-069, rev. 5., April 1996.

[CAO-5]
"Transuranic Waste Characterization Quality Assurance Program Plan," United States Department of Energy Carlsbad Area Office, CAO-94-1010, rev. 0., *Interim Change*, November 15, 1996.

[CAO-6]
"WIPP RCRA Part B Permit Application, Appendix C9, TRU Waste Characterization Using Acceptable Knowledge," United States Department of Energy Carlsbad Area Office, DOE/WIPP 91-005, rev. 6., 1996.

[CAO-7]
"Title 40 CFR 191 Compliance Certification Application for the Waste Isolation Pilot Plant, Appendix WCL" United States Department of Energy Carlsbad Area Office, DOE/CAO 1996-2184, October 1996.

[CAO-8]
"Transuranic Waste Baseline Inventory Report," United States Department of Energy Carlsbad Area Office, DOE/CAO-95-1121, rev. 3., June 1996.

[CAO-9]
"Performance Demonstration Program Plan for RCRA Constituent Analysis of Solidified Wastes", Rev. 1, DOE/CAO-95-1077, United States Department of Energy Carlsbad Area Office, March 1996.

[CAO-10]
"Performance Demonstration Program for Nondestructive Assay for the TRU Waste Characterization Program, Scoring Report, April 1996 Distribution (Cycle 1)," prepared by Lockheed Martin Idaho Technologies Company for the US. DOE Carlsbad Area Office, CAO:NTP:JFS 96-1579, July 1996.

[CAO-11]
"Performance Demonstration Program for Nondestructive Assay for the TRU Waste Characterization Program, Scoring Report, November 1996 Distribution (Cycle 2)," prepared by Lockheed Martin Idaho Technologies Company for the US. DOE Carlsbad Area Office, CAO:NTP:JFS 97-0223 UFC 5822, January 1997.

[CAO-12]
"Performance Demonstration Program Plan for Nondestructive Assay for the TRU Waste Characterization Program", U.S. DOE, Carlsbad Area Office, CAO-94-1045, rev. 1, May 1997.

[DOE-1]
"Radioactive Waste Management," DOE Order 5820.2A. September 26, 1988.

[DOE-2]
"Internal Dose Conversion Factors for Calculation of Dose to the Public," U.S. Department of Energy, DOE/EH-0071, July 1988.

[DOE-3]
"DOE Methods for Evaluating Environmental and Waste Management Samples", U.S. Department of Energy Assistant Secretary for Environmental Restoration and Waste Management, DOE/EM-0089T, Rev.2.

[DOE-4]
Multianalyte Performance Evaluation Program, U.S. Department of Energy, Idaho Operations Office, 850 Energy Drive, Idaho, Falls, Idaho 83401-1563.

[DOE-EM-1]
Grumbly T.P. "Institutionalizing the Data Quality Objectives Process for EM's Environmental Data Collection Activities," United States Department of Energy Memorandum, EM-263, {http://etd.pnl.gov:2080/DQO/memo/grumbly.html}, September 7, 1996.

[DOE-EM-2]
"Data Quality Objectives," website operated for the U. S. Department of Energy Office of Environmental Management (EM-76) by the Pacific Northwest National Laboratory, http://etd.pnl.gov:2080/cgi-bin/DQOhome.cgi.

[EML-1]
"Environmental Measurements Procedures Manual", HASL-300, U.S. Department Of Energy, New York, NY.

[EML-2]
Environmental Measurements Laboratory Quality Assessment Program, U.S. Department of Energy Environmental Measurements Laboratory, Office of Environmental Management, New York, NY 10014-3621.

[EPA-1]
"Guidance for Planning for Data Collection in Support of Environmental Decision Making Using the Data Quality Objectives Process," EPA/QA/G-4, United States Environmental Protection Agency, September, 1994.

[EPA-2]
"Test Methods for Evaluating Solid Waste", SW-846, 3rd ed., Office of Solid Waste and Emergency Response, Washington, D. C., Nov. 1986; Update I, July 1992, and Final Update II, Sept. 1994.

[EPA-3]
"Methods for Chemical Analysis of Water and Wastes", U.S. Environmental Protection Agency, Environmental Monitoring and Support Laboratory, EPA-600/4-79-020, current revision.

[IAEA-1]
"Hazardous Materials, Transportation Regulations; Compatibility with Regulations of the International Atomic Energy Agency," HM-169A, 1996.

[INEEL-1]
"5th Nondestructive Assay and Nondestructive Examination Waste Characterization Conference," Conference Proceedings, Salt Lake City, UT, January 14-16, 1997.

[INEEL-2]
"4th Nondestructive Assay and Nondestructive Examination Waste Characterization Conference," *conference proceedings*, Salt Lake City, UT, October 24-26, 1995.

[INEEL-3]
"Modelling Parameters for RH-TRU 30-gallon Drums-HKP-02-96," Interdepartmental Communication from H. Peterson to Y. Harker, March 13, 1996.

[INEEL-4]
"Alpha Gamma Hot Cell Facility Operations Manual", Section 11, IPS-2-00-00, Rev. 2, 9/10/90, Argonne National Laboratory.

[INEEL-5]
Hartwell, J.K., Yoon, W.Y., and Peterson, H.K., "A Preliminary Evaluation of Certain NDA Techniques for RH-TRU Characterization", Idaho National Engineering and Environmental Laboratory, Proceedings, 5th Nondestructive Assay and Nondestructive Examination Waste Characterization Conference, Salt Lake City, Utah, January 14-16, 1997.

[INEEL-6]
Tyler, C.R., Gladson S.C., Ebner, M.A., Cole, J.D., Mandler, J.W., Peterson, H.K. "Joint Program Development of a Direct Measurement Nondestructive Assay System for RH-TRU Waste Using GNAT/FATS Technology," Lockheed Martin Idaho Technologies Company, Nov. 20, 1996.

[LANL-1]
Coop K.L., Estep R.J. "Description of the PAN Assay System for Remote-Handled Waste," LA-UR-91-125, Los Alamos National Laboratory, January 7, 1991.

[LANL-2]
Estep R.J., Coop K.L., Deane T.M., Lujan J.E., "A Passive-Active Neutron Device for Assaying Remote-Handled Transuranic Waste," LA-UR-89-3736, Los Alamos National Laboratory, November 17, 1989.

[LANL-3]

N. J. Nicholas, K. L. Coop, and R. J. Estep, "Capability and Limitation Study of the DDT Passive-Active Neutron Waste Instrument," Los Alamos National Laboratory report LA-12237-MS, April, 1992.

[NUREG-1]
Reilly D., Ensslin N., Smith H. (ed.), "Passive Nondestructive Assay of Nuclear Materials," NUREG/CR-5550, LA-UR-90-732, Office of Nuclear Regulatory Research, USNRC, March 1991.

[NUREG-2]
Gozani, T , "Active Nondestructive Assay of Nuclear Materials, Principles and Applications" NUREG/CR-0602, January 1981.

[ORNL-1]
Stewart R.C., "Oak Ridge National Laboratory, Remote-Handled Transuranic Solid Waste Characterization Study," Oak Ridge National Laboratory, ORNL/TM-11050, June 1989.

[ORNL-2]
Nguyen L., Yong L., Chapman J., Beachamp J., Downing D., Federov V., Parks C., Schreiber R., Schultz F. "Project Plan for the Evaluation of REDC Waste for TRU-Waste Radionuclides," Oak Ridge National Laboratory, ORNL/TM-13087, September 1996.

[ORNL-3]
Beachamp J., Downing D., Chapman J., Federov V., Nguyen L., Parks C., Schultz F., Yong L. "Statistical Analysis of Radiochemical Measurements of TRU Radionuclides in REDC Waste," Oak Ridge National Laboratory, ORNL/TM-13298, October 1996.

[ORNL-4]
"Radiofrequency Quadrupole Assay System Test Project On-site RFQ Assay Equipment Operability Test Milestone and the Operational Readiness Process," SM. Stoller Corporation report for Oak Ridge National Laboratory, September 30, 1996.

[ORNL-5]
"Project Management Plan for the Radiofrequency Quadrupole Assay System Test Project at the Oak Ridge National Laboratory, WMRA-ARM-809, SM. Stoller Corporation report for Oak Ridge National Laboratory, September 30, 1996.

[ORNL-6]
"A Workshop on Characterizing Transuranic Waste," sponsored by the Applied Radiation Measurements Department, Oak Ridge National Laboratory, August 16, 1995.

[ORNL-7]
Martin R., "Estimating Source Terms Using the ORIGEN2 Computer Code, " presented at the Workshop on Characterizing Transuranic Waste," sponsored by the Applied Radiation Measurements Department, Oak Ridge National Laboratory, August 16, 1995.

[ORNL-8]

Benker D.,"The REDC Chemical Separations Process, " presented at the Workshop on Characterizing Transuranic Waste," sponsored by the Applied Radiation Measurements Department, Oak Ridge National Laboratory, August 16, 1995.

[ORNL-9]
Schreiber D.,"Characterization of Radioactive Contamination in Waste at REDC, " presented at the Workshop on Characterizing Transuranic Waste," sponsored by the Applied Radiation Measurements Department, Oak Ridge National Laboratory, August 16, 1995.

[ORNL-10]
Schultz F.J., Vourvopoulos G., Womble P.C., Roberts M.L., "A Feasibility Study for a LINAC-based Transuranic Waste Characterization System," *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 193, No. 2 (1995).

[ORNL-11]
DeVore, J.R., et.al., "Statistical Analysis of ORNL LLLW System Transuranic Wastes", ORNL/TM-13351, Oak Ridge National Laboratory, Oak Ridge, TN, Dec. 1996.

[ORNL-12]
Keller, J.M., Giaquinto, J.M., Meeks, A.M., "Characterization of the MVST Waste Tanks Located at ORNL", ORNL/TM-13357, Oak Ridge National Laboratory, Oak Ridge, TN, December, 1996.

[ORNL-13]
Keller, J.M., Giaquinto, J.M., Meeks, A.M., "Characterization of the Old Hydrofracture (OHF) Waste Tanks Located at ORNL", ORNL/TM-13394, Oak Ridge National Laboratory, Oak Ridge, TN, April, 1997.

[ORNL-14]
Keller, J.M., Giaquinto, J.M., Meeks, A.M., "Characterization of the BVEST Waste Tanks Located at ORNL", ORNL/TM-13358, Oak Ridge National Laboratory, Oak Ridge, TN, January, 1997.

[ORNL-15]
"Radioactive Materials Analysis Laboratory-Oak Ridge National Laboratory Quality Assurance Project Plan for the Transuranic Waste Characterization Program", QAP-X-94-CASD/RML-002, rev. 2, Oak Ridge National Laboratory, May 1996.

[ORNL-16]
Nguyen, L.K., Miller, L.F., Downing, D.J. "Measurements of Fission and Activation Products for Oak Ridge National Laboratory Waste Characterization," ORNL/TM-13292, June 1997.

[ORNL-17]
Womble, P.C., Vourvopoulos, G.V., Ball, J.C., Persons, K.A. "Calculations of Neutron Transport Through a Simulated Waste Matrix, " submitted and accepted to *Nucl. Instr. Meth. In Phys. Res.*, in press as of July 1997.

[SAND-1]

"Radioisotopes to be used in the 1996 CCA Calculations," internal memo from J.W. Garner to C.T. Stockman, Sandia National Laboratories, March 15, 1996.

[SAND-2]
"Concerns on QAOs Criteria," internal memo from L. C. Sanchez to D. R. Anderson, Sandia National Laboratories, December 7, 1995.

[SAND-3]
"Updated (2033yr) Radionuclides For Use in WIPP PA Calculations,"internal memo from L. C. Sanchez to M. Martell, Sandia National Laboratories, October 28, 1996.

[SAND-4]
"Identification of Important Radionuclides Used in 1996 CCA WIPP Performance Assessment," internal memo from L. C. Sanchez to distribution, Sandia National Laboratories, April 25, 1996.

[SAND-5]
Sanchez, L.C., Liscum-Powell, J., Rath, J.S., Trellue, H.R., "WIPP PA Analysis Report for EPAUNI: Estimating Probability for Performance Assessment Calculation", Document Version 1.01, WBS# 1.2.07.1.1, February 17, 1997.

[WEST-1]
"Safety Analysis Report for Packaging for the RH-TRU 72B Waste Shipping Package," Westinghouse Corp., April 1995. {rev. 0 released by Vectra Technologes, Dec. 1994}

7.0 APPENDICES

APPENDIX A: WIPP PA ANALYSIS REPORT SUMMARY TABLES

Section 2.2.1 of this report provides a summary of the WIPP Performance Assessment-based regulatory drivers for the management of TRU waste. Table 2.3 of that section breaks out the significant isotopic differences between reported CH and RH inventories. To help clarify table 2.3, the following tables were taken directly from reference SAND-5 and are a partial representation of the data tables available in SAND-5 indicating the relationship between the projected TRU inventory and EPA normalized release units. Refer to SAND-5 for more detailed information.

Table 7.1 lists the five most significant radionuclides, expressed as a cumulative percentage of an EPA-regulated waste unit. Note that the percent of total activity for a given isotope (shown in table 2.3) is not equal to the Cum. % of Unit of Waste (shown in Table 7.1). As defined in 40CFR191, the EPA regulates WIPP for radioactivity on the basis of an EPA unit and not on the basis of total activity. Combining the data of table 2.3 with the data in 7.1, one observes the RH-TRU waste has a less than 0.1% impact on repository performance. This is the significance of the results reported in SAND-5.

TABLE 7.1

40CFR191 Unit of Waste for WIPP-Scale TRU Waste (a) Top 5 Radionuclides			
Nuclide ID	**Transuranic Inventory [α-Curies](b)**		**Cum. % of Unit of Waste**
	1995	2033	
^{238}Pu	2.61E+06	1.94E+06	56.4
^{239}Pu	7.96E+05	7.95E+05	79.5
^{241}Am	4.48E+05	4.88E+05	93.7
^{240}Pu	2.15E+05	2.14E+05	99.9
^{242}Pu	1.17E+03	1.17E+03	>99.9
	Sum =	3.44E+06 (WUF =3.44)	

Table 7.2 provides a little more detail on the top 10 nuclides, with the major difference being that contribution from ^{137}Cs and ^{90}Sr. The values in tables 7.1 and 7.2 may appear to be inconsistent, but the only difference is when the RH contribution from the major fission products is included. Using only 4 radionuclides (^{238}Pu, ^{239}Pu, ^{241}Am, ^{240}Pu) 99.4% of the EPA unit is accounted for. Using 7 radionuclides (adding ^{137}Cs and ^{90}Sr) accounts for greater than 99.9% of the EPA unit. Table 7.2 allows one to intercompare the differences in inventory between CH and RH waste. There is no significant contribution to the EPA unit from RH waste.

TABLE 7.2

40CFR191 Release Limits and Source Term EPA Units for WIPP-Scale TRU Waste (Calendar Year = 2033, Time = 0 yr) (a) Top 10 Radionuclides							
ID	**Total Inventory (Curies)**			**Source Term EPA Unit (e)**			
	CH	**RH**	**Total**	**CH**	**RH**	**Total**	**Cum (%)**
^{238}Pu	1.93E+06 (c)	1.08E+03	1.94E+06	5.61E+03	3.14E+00	5.64E+03	56.0
^{239}Pu	7.85E+05	1.03E+04	7.95E+05	2.28E+03	2.99E+01	2.31E+03	79.0
^{241}Am	4.78E+05	9.43E+03	4.88E+05	1.39E+03	2.74E+01	1.42E+03	93.2
^{240}Pu	2.09E+05	5.05E+03	2.14E+05	6.08E+02	1.47E+01	6.22E+02	99.4
^{137}Cs	3.35E+03	8.98E+04 (d)	9.31E+04	9.74E+01	2.61E+01	2.71E+01	99.6
^{90}Sr	2.77E+03	8.45E+04	8.73E+04	8.05E+01	2.46E+01	2.54E+01	99.9
^{233}U	1.79E+03	1.58E+02	1.95E+03	5.20E+00	4.59E+01	5.67E+00	>99.9
^{242}Pu	1.17E+03	1.50E+01	1.17E+03	3.40E+00	4.36E+04	3.40E+00	>99.9
^{243}U	7.08E+02	4.29E+01	7.51E+02	2.06E+00	1.25E+01	2.18E+00	>99.9
^{237}Np	6.19E+01	2.95E+00	6.49E+01	1.80E+01	8.58E+08	1.89E+01	>99.9

(a) Radionuclide inventory information taken from Transuranic Waste Baseline Inventory Database (TWBID), Revision 3, [CAO-8]. (In total, 135 radionuclides are inventoried in the TWBID.)

(b) Transuranic inventory (curie) data taken from Ref. [CAO-8] . Values correspond to a "WIPP-Scale" design basis.

(c) Isotope with dominant curie load for CH-TRU wastes.

(d) Isotope with dominant curie load for RH-TRU wastes.

(e) The "Source Term EPA Unit" is the Inventory (CH, RH, Total) divided by the 40CFR191 App A. Release Limit (Ci/UW) times the WUF. For example, for ^{238}Pu the CH inventory is 1.93E+06 Ci. The 40CFR191 release limit is 11 Ci/UW. The WUF is 3.44. The CH Source Term EPA unit is thus 1.93E+06/(100*3.44) = 5.61E+03.

Other notes of interest from SAND-5 include:

– The total CH-TRU curie load is 3.80 MCi (also, 90.6% of the total curie load in WIPP) at the year 2033, as reported in other tables in ref. SAND-5. The average CH-TRU volumetric total curie load is 3.80E+06/5.95E+06 = 0.639 (Ci/cu. ft.) = 22.6 (CVcu. m.).[CH volume in SAND-5 -- →5.95E+06 cu. ft. Also note that the 5.95E+06 volume number is a volume limit, stored waste and projected waste volumes sum up to a volume less than this volume. The summed up volume is then "scaled" to yield a total volume of 5.95E+06, likewise the curie loads are also "scaled" by the same factor. Thus ratios of curie loads (or heat loads) to volume yield values are applicable to expected waste at WIPP.]

- The total RH-TRU curie load is only 0.393 MCi (also, only 9.37 % of the total curie load in WIPP) at the year 2033, as reported in other tables in ref. SAND-5. This curie load is much less than the RH-TRU limit of 5.1 MCi. The average RH-TRU volumetric total curie load is 0.393E+06/0.25E+06 = 1.57 (Ci/cu. ft.) = 55.5 (Ci/cu. m.). [RH volume in SAND-5 --→0.25E+06 cu. ft.]

- There are only 16 radionuclides in the TWBIR that comprise the “transuranic waste”. Eleven of these radionuclides are included in the WIPP PA data base and correspond to 99.999% of the total RH “transuranic” inventory. The average RH-TRU volumetric “transuranic” curie load is 2.59E+04/0.25E+06 = 0.104 (TRU-Cl/cu. ft.) = 3.66 (TRU-Cl/cu. m.). [The RH contribution to the WIPP-scale inventory of transuranic curie load is 2.59E+04*100%(2.59E+04 + 3.41E+06) = 0.75% (very small).] Note, this value is an order of magnitude less than that presented for the CH-TRU wastes -- since the majority of the curie content of the RH-TRU waste is due to shorter-lived non-transuranic radionuclides, this means that as the shorter-lived components decay away the remaining RH-TRU waste will have a lesser curie content than CH-TRU waste. Since the WIPP Passive Institutional Controls (PICs) are credited for several hundred years after the closure of WIPP, it is evident that RH-TRU does not have a significant effect on performance assessment (PA) of WIPP.

APPENDIX B: DESCRIPTION OF THE OAK RIDGE ISOTOPE GENERATION CODE

Isotopes known as fission products result from fission, that process whereby a fissionable nucleus splits following the interaction by a neutron. In essence, the nucleus splits into two nuclei accompanied by the emission of several neutrons and gamma-rays. The two nuclei that emerge from the reaction are fission products, some with very short half-lives, and others up to about 30 years. Fission is a statistical process; the two fission nuclei that are produced from any two fission events are not likely to be the same. A distribution of fission-product nuclei will emerge after enough fissions are observed. It is this fission-product distribution and corresponding magnitude that is computed by ORIGEN. These distributions are very dependent on reactor conditions, the type of fuel, and neutron moderation, but easily accounted for by well validated computer codes. For example, figure 7.1 shows the nominal fission-product distribution that results from the fission of ^{235}U achieved by thermal neutron irradiation. The commonly encountered fission products, ^{90}Sr and ^{137}Cs, are maxima in the bimodal distribution. Thus it should be no surprise that a large fraction of the RH-TRU waste generated from the nuclear fuel cycle contains a significant quantity of ^{137}Cs and ^{90}Sr. Many of the reactor fuels or targets that were tested in the DOE programs---whose waste materials are being managed as RH-TRU waste---did not consist of ^{235}U, or were not fissioned by thermal neutrons. Subsequently, outputs from ORIGEN runs for RH-TRU waste may look quite a bit different between waste generators. For example, the RH-TRU waste originating in Oak Ridge is largely produced as a result of Cm-target irradiation in the High Flux Isotope Reactor. [ORNL-7] Two types of targets have been used. The figure shows that the fission-product distribution for this process is quite different than for fuel bearing ^{235}U, as expected. ORIGEN data will provide a sound basis for process knowledge in the management of RH-TRU, but like anything else, has advantages and limitations as described below.

Advantages:

1. Provides lower and upper limits on the amount of radioactivity produced over an entire waste-generating process;
2. Provides lower and upper limits on the amount of radioactivity produced over separate campaigns of an entire process, provided the campaigns are truly independent from one another, (e.g. minimal cross contamination between campaigns or batches);
3. Allows some isotopes to be excluded from consideration for measuring and reporting, (e.g. the production fractions of some isotopes, while not zero, are so low that they are of no consequence to RH waste management);
4. Provides baseline isotopic proportions;
5. Decays radioisotopic activity with the passing of time, which is important in most cases, where the waste will not be certified for WIPP until after the year 2001.

Limitations:

1. Not necessarily adaptable for computing distributions of transuranium products in fuel or targets. To compute transuranium formation in curium targets, ORNL uses a code called TCOMP, for example.

2. Cannot account for complicated processes that smear isotopes between batches or that separate isotopes from one another by chemical methods, as suggested in §4.2. When these processes occur, the advantages of ORIGEN are reduced essentially to 1 and 4 listed previously.

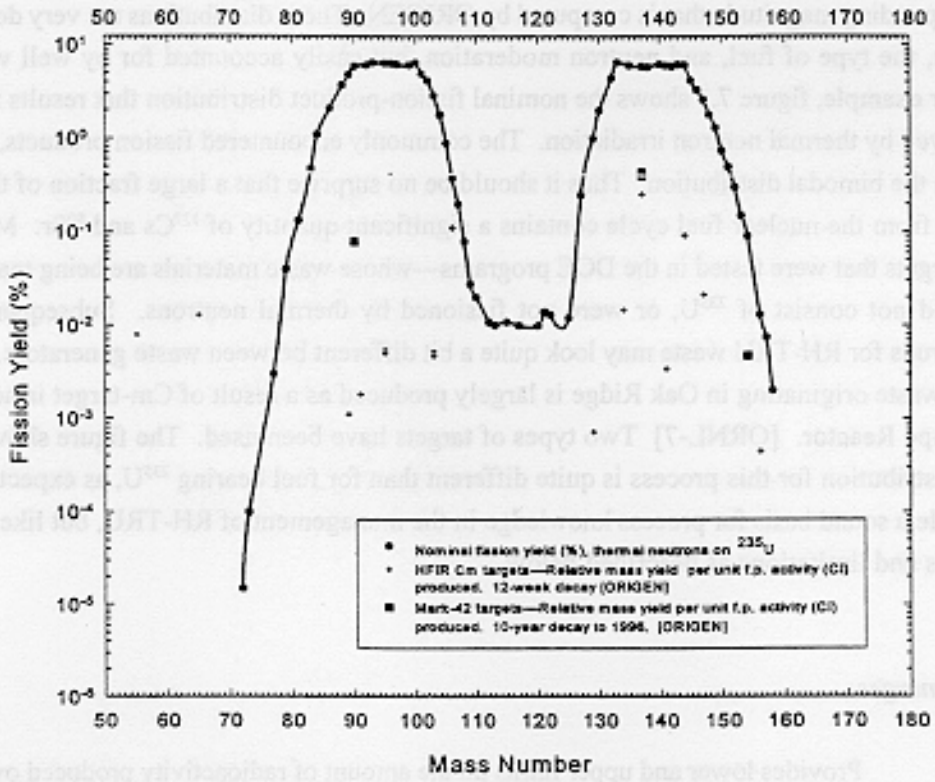

Figure 7.1 Fission Product Distribution for Thermal Neutron Fission of ^{235}U.
Curium target data from the HFIR superimposed to show relative difference in fission-product distribution. The Cm target data is not normalized per fission, but rather, per curie of fission-product activity produced.

APPENDIX C: NONDESTRUCTIVE ASSAY OF CONTACT-HANDLED WASTE

Implementation of nondestructive assay (NDA) techniques for the characterization of DOE contact-handled transuranic (CH-TRU) waste stemmed from NDA development activities funded by the DOE Safeguards and Security Office. These programs fostered the development of reliable, non-intrusive NDA methodologies to confirm inventories of special nuclear material (SNM); in particular, plutonium. Both passive and active neutron and gamma-ray techniques were developed and implemented and included passive neutron coincidence counting (PNCC), active well coincidence counter (AWCC), various transmission-corrected gamma-ray techniques, and so-called passive-active neutron techniques (e.g., ^{252}Cf Shuffler and Differential Dieaway Technique, DDT).

In the 1970's, some of these techniques were first applied to the characterization of CH-TRU waste; in particular, transmission-corrected gamma-ray and DDT. The application of transmission-corrected gamma-ray methodologies developed into a technique called Segmented Gamma Scanner (SGS), while the application of DDT developed into a technique eventually called Passive Active Neutron (PAN). The particular challenges the characterization of CH-TRU waste presented the scientific community were not trivial. The characterization of SNM was a simpler endeavor since the SNM was usually well-characterized by other means and the NDA measurements were simply a confirmatory measurement to ensure safeguards requirements had not been violated. Also, the SNM was typically homogeneous and uniformly distributed within its container. On the other hand, characterization of TRU waste is more complex. For example, often the contents of the waste container are unknown and, consequently, the NDA measurement must provide primary characterization data rather than confirm previous data. Further complicating the measurements is the nature of the waste itself; that is, heterogeneous matrices and non-uniformly distributed radioactivity. Both of these features present significant challenges to any NDA waste measurement protocol. The key to successful implementation of these methodologies is diligent calibration combined with good engineering practice to assure that the instrument is being applied within the calibration envelope. Two NDA techniques which have been extensively used in characterizing CH-TRU waste, SGS and PAN, will be discussed in some detail.

The SGS technique [App.C ref., Aug74], when applied to the characterization of CH-TRU waste contained in drums (typically, 55-gallon), involves, first, the detection of gamma rays emitted by the TRU waste radionuclides (see DOE Order 5820.2A) and fission and activation products. A selected list of radionuclides and their corresponding gamma-ray energies is provided in Table 7.3. Second, the intensities of the gamma rays of interest (i.e., photons per unit time detected) are corrected by appropriate attenuation factors. These factors are determined by the relative intensities of known gamma rays after passage through air, an empty drum, and the waste drum, respectively. For example, a selenium-75 source (gamma-ray energy equal to 400.6 keV) is used to correct for the attenuation that a ^{239}Pu 414 keV gamma ray suffers as it traverses through the waste matrix, drum wall, and air to the gamma-ray detector. Examples of a weapons-grade ^{239}Pu gamma-ray spectrum and a spectrum obtained from the measurement of actual ORNL RH-TRU waste obtained using a SGS system is provided in Figure 7.1.

The PAN system was developed at Los Alamos National Laboratory in the late 1970s [App. C ref., Kun81] and was awarded the 1983 IR-100 award. [App. C ref., Nic92] It was first developed to sort waste, by bulk measurement, into two categories: low-level and TRU. At that time, TRU waste was defined as waste bearing more than 10 nCi/g of transuranium radionuclides, predominantly ^{239}Pu, ^{240}Pu, and ^{241}Am. The active portion of PAN, i.e., DDT, was developed to preferentially measure fissile material, ^{239}Pu and ^{235}U. Under some conditions, it can measure fissile material in a 55-gallon drum down to milligram quantities. Kunz initially reported that the sensitivity of the system was 0.9 mg ^{239}Pu and 1.3 mg ^{235}U, for an assay time of 100 sec (10^4 generator pulses at 100 cycles per second).[App. C ref., Kun81] DDT cannot distinguish between ^{239}Pu and ^{235}U, but is more sensitive to ^{239}Pu by a factor of 1.5. An excellent overview of the method is found in [App. C ref., Sch91].

The DDT method is straightforward, in principle. It directly measures fissile material (e.g., ^{239}Pu, ^{235}U) by causing a small percentage of the fissile mass to fission and then detects the resulting fast-fission neutrons released during the fission process. A pulsed-neutron generator provides the source of neutrons for inducing the fissions. A typical neutron generator produces 10^6 14-MeV neutrons per pulse and is pulsed at a rate of 50 cycles per second. Typically, for each measurement, the generator is operated for 40 seconds, which translates to 2000 pulses.

Between each pulse, the neutrons introduced from the generator scatter in the chamber and in the drum, eventually reaching very low energies at which point they can readily be captured by a fraction of the fissile material present, resulting in fission. Each fission produces 2.5 neutrons on average, which, much like the interrogating neutrons, scatter inside the drum, then emerge, and are detected in an array of neutron detectors. In effect, the neutron generator is turned on for 10 µsec, every 20 msec. While the generator is off, the system neutron detectors are turned on to detect the neutrons resulting from induced fission. This process is repeated 2000 times in 40 seconds to achieve a very low level of detection.

One way of visualizing this process is to observe data for two separate cases: with and without ^{235}U present. Figure 7.2 shows a typical differential dieaway time semi-log spectrum for a system with and without ^{235}U present. Note that the time duration of the multi-channel spectrum is 20 msec (20,000 µsec). For the lower curve, from time zero to 600-700 µsec the initial burst of neutrons from the generator "dies away" exponentially. After 600-700 µsec, the only neutrons detected are due to natural background - cosmic ray interactions with nearby materials. The background rate is approximately 4 to 6 per 10-µsec wide channel. Notice the large difference in the magnitude of the peak neutron rate from the generator to that rate from background neutrons alone. This phenomenon is part of the reason why DDT is very sensitive to the measurement of ^{235}U. It introduces a large potential signal relative to noise.

The upper curve of figure 7.2 depicts a differential dieaway time spectrum with ^{235}U present. The number of neutrons detected between the time interval from about 600 µsec to 4700 µsec is dramatically increased over the background case. The amount of fissile material present is proportional to the number of neutrons detected during this time interval. The mass of ^{235}U present is proportional to the net number of neutrons detected, normalized by several neutron monitors that account for differences between the neutron flux produced from the

neutron generator and the neutron flux resulting from induced fission. Other corrections are usually made to account for matrix-specific effects, such as neutron moderation and absorption. These effects can diminish and/or distort the signal.

Both the SGS and DDT methods have been continuously studied and improvements have been made to each. For example. Combined Thermal Epithermal Neutron (CTEN) [App. C ref., Coop89], Imaging Passive Active Neutron (IPAN) [App. C ref., Cald91], and Active-Passive Neutron Examination and Assay (APNea) [App. C ref., Hen95a, Hen95b] are variations of the original PAN/DDT systems. Tomographic Gamma Scanner (TGS) [App. C ref., Estep94], Active and Passive Computerized Tomography (A&PCT) [App. C ref., Camp91], [App. C ref., Rob95], are variations of the original SGS systems.

Table 7.3. Selected radionuclides and their corresponding gamma-ray energies

Radionuclide	Gamma-ray Energy, keV
^{235}U	185
^{239}Pu	414
^{241}Am	59
^{137}Cs	662
^{60}Co	1173 and 1332

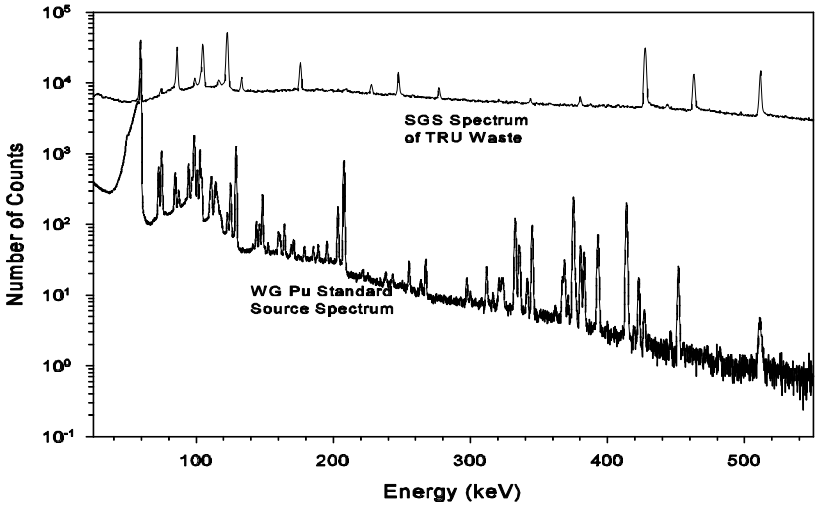

Figure 7.1. **Gamma-ray spectra** (A conventional, well-resolved Pu spectrum of CH-TRU waste is compared against an RH-TRU spectrum from ORNL. In the RH-TRU spectrum, no TRU photopeaks are resolved above the compton continuum.)

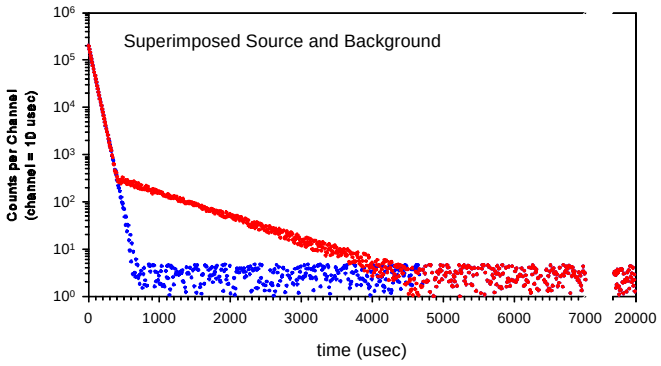

Figure 7.2 Time spectra from background and source are superimposed to show the relative magnitude of signal to noise in a DDT system.

Appendix C References

[Aug74]

Augustson, R. H. and Reilly, T. D., "Fundamentals of Passive Nondestructive Assay of Fissionable Material," LA-5651-M, September 1974.

[Coop89]

Coop, K. L., "A Combined Thermal/Epithermal Neutron Interrogation Device to Assay Fissile Material in Large Containers," presented at the 11th Annual ESARDA Symposium on Safeguards and Nuclear Material Management European Centre (New Hemicycle), Luxembourg, June 1989, LA-UR-89-1685.

[Cald91]

Caldwell, J. T., et al., "Greatly Improved Transuranic Waste Assay Accuracy Using Neutron Signal Imaging," invited paper presented at the ANS 1991 Meeting, San Francisco, CA, November 10-14, 1991, Vol 64, TANSAO 64 1-754 (1991).

[Camp91]

Camp, D. C. and Martz, H. E., "Nondestructive and Quantitative Characterization of TRU and LLW Mixed-Waste Using Active and Passive Gamma-Ray Spectrometry and Computed Tomography," UCRL-ID-108882.

[Rob95]

Roberson, G. P., et al., "Nondestructive Assay of TRU Waste Using Gamma-Ray Active and Passive Computed Tomography," paper presented at the 4th Nondestructive Assay and Nondestructive Examination Waste Characterization Conference, Salt Lake City, UT, October 24-26, 1995, CONF-951091.

[Estep94]

Estep, R. J., et al., "Tomographic Gamma Scanning to Assay Heterogeneous Radioactive Waste," Nucl. Sci. Eng., 118, 145 (1994).

APPENDIX D: DESCRIPTION OF ANALYTICAL INSTRUMENTATION TYPICALLY USED FOR DESTRUCTIVE RADIOASSAY

[ref. APHA-1]

Alpha Spectrometers

Alpha spectrometers are semiconductor particle detectors which measure alpha emitters based upon the discrete energy of the alpha emitter. There are primarily two types of these detectors and both are silicon based; the silicon surface barrier detector or the passivated implanted planar silicon detector (PIPS). Typically, sample preparation for alpha spectrometry involves chemical separation of the sample to remove sample matrix and spectroscopic interferences followed by evaporation or electrodeposition onto a counting planchet. Subsequently alpha emissions from a sample enter the detector and interact with detector atoms. This results in ionization within the detector and collection of the charged particles via a bias voltage applied to the detector. Consequently, an electrical voltage pulse occurs which is proportional to the energy of the deposited alpha particle. Amplifiers, analog-to-digital converters, and analyzers are used to convert the electrical signals into spectra and activity counts. The detector performance is primarily affected by resolution, active area, and depletion depth.

Geiger-Mueller and Proportional Counters

Geiger-Mueller and proportional counters are used for gross measurements of beta and alpha activity. Typically, gas flow proportional counters are used for radioassay due to better counting efficiencies. Samples are deposited via evaporation or electrodeposition onto a counting planchet and placed in counting shelves within a counting chamber. Radiation is then emitted into a counting gas, which in turn is ionized resulting in electrons being deposited onto an anode of the counting chamber. Signals are then amplified and counts are recorded. Counts are converted to activity via efficiency calculations using known standards (efficiency ~ counts/decay).

Gamma Spectrometers

Two primary types of gamma spectrometers are in wide use: (1) the sodium iodide, thallium-activated {NaI(Tl)} crystal system using a scintillation phenomenon and (2) high purity germanium detectors {HP(Ge)}using semiconductor technology. Gamma spectrometers allow quantification of individual gamma emitters based upon the energy of the gamma emitter. The NaI(Tl) detectors are based upon the production of light pulses from interaction of gamma photons with the scintillation material. The light pulses are detected with a photomultiplier tube which converts the light pulses to electronic voltage pulses proportional to the light intensity. Signals are then amplified, spectra are produced, and counts are recorded. Germanium detectors are based upon the "ionization" of atoms within a semiconductor crystal as the gamma photons deposit energy into the crystal. This ionization creates the movement of electrons throughout the crystal which creates an electrical signal proportional

to the deposited photon energy. Signals are then amplified, spectra are produced, and counts are recorded. Germanium detectors have higher resolution than do sodium iodide detectors while the NaI(Tl) detectors have a much better efficiency. Minimal sample preparation is involved with either type of system; the sample must simply be adapted to a geometry consistent with the geometry of the standard used to calibrate the system.

Liquid Scintillation Counters

Liquid scintillation counters are used for measuring gross beta and gross alpha activity. This technique is based upon the use of a liquid scintillator which is added to the sample. The radiation in the sample interacts with the scintillator resulting in flashes of light. The light flashes are then detected and amplified by one or more photomultiplier tubes and counts are recorded. Sample preparation involves dissolving or suspending the sample in a liquid scintillation solution and placing in a translucent bottle to enable light flashes to be transmitted to the photomultiplier tubes.

APPENDIX E: OAK RIDGE NATIONAL LABORATORY SLUDGE DATA TABLES

Table 7.4. Estimates for ^{239}Pu FGE with the ORNL MVST Sludge

Isotope	^{239}Pu FGE factor	Tank W-24 (mg/Kg)	Tank W-25 (mg/Kg)	Tank W-26 (mg/Kg)	Tank W-27 (mg/Kg)	Tank W-28 (mg/Kg)	Tank W-31 (mg/Kg)
^{233}U	0.865	3.58	6.6	25.1	2.52	12.0	10.9
^{235}U	0.641	33.2	41.0	51.3	35.7	46.2	121
^{239}Pu	1.000	0.84	1.46	0.57	0.29	0.36	1.49
^{239}Pu FGE (mg/Kg)		25.22	33.45	55.16	25.35	40.35	88.48
^{239}Pu FGE in 55 gal. (g)		**7.2**	**9.5**	**15.8**	**7.6**	**11.5**	**26.5**
Percent of WAC (325g) per 3 drums		**7%**	**9%**	**15%**	**7%**	**11%**	**24%**

Table 7.5 Estimates for ^{239}Pu Equivalent Activity with the ORNL MVST Sludge

Isotope	^{239}Pu wt. factor[a]	Tank W-24 (Bq/g)	Tank W-25 (Bq/g)	Tank W-26 (Bq/g)	Tank W-27 (Bq/g)	Tank W-28 (Bq/g)	Tank W-31 (Bq/g)
^{233}U	3.9	1600	2800	10000	1000	5200	5200
^{238}Pu	1.1	3800	7800	5400	2400	3000	13000
^{239}Pu	1.0	1900	3400	1300	670	830	3400
^{240}Pu	1.0	870	1800	890	370	600	2200
^{241}Pu	52.0	14000	26000	15000	6500	12000	24000
^{241}Am	1.0	3900	9300	3900	2800	4600	14000
^{244}Cm	1.9	22000	58000	28000	17000	25000	110000
^{239}Pu Eqv. (Bq/g)		22382.98	53335.17	28588.50	15350.60	23479.27	91107.79
^{239}Pu Eqv. in 55 gal. (Ci)		**0.17**	**0.41**	**0.22**	**0.12**	**0.18**	**0.74**
Percent of WAC (1000Ci) per 3 drums		**0.1%**	**0.1%**	**0.1%**	**0.1%**	**0.1%**	**0.2%**

[a] Radionuclide-specific weighting factors for the ^{239}Pu equivalent activity taken from Appendix A of DOE/WIPP-069, Rev.5

Table 7.6 Isotopes that Contribute to the Decay Heat in the MVST Sludge

Isotope	"Q" value (W/Ci)	"Q" value (W/Bq)	Tank W-24 (Bq/g)	Tank W-25 (Bq/g)	Tank W-26 (Bq/g)	Tank W-27 (Bq/g)	Tank W-28 (Bq/g)	Tank W-31 (Bq/g)
^{60}Co	1.54E-02	4.16E-13	2.80E+04	2.50E+04	5.80E+04	1.20E+04	4.20E+04	2.20E+04
^{90}Sr	1.16E-03	3.14E-14	1.40E+06	3.20E+06	7.10E+05	4.50E+05	7.00E+05	1.10E+07
^{90}Y	5.54E-03	1.50E-13	1.40E+06	3.20E+06	7.10E+05	4.50E+05	7.00E+05	1.10E+07
^{137}Cs	1.01E-03	2.73E-14	5.30E+05	4.70E+05	8.90E+05	3.90E+05	3.10E+05	4.30E+05
^{137m}Ba	3.94E-03	1.06E-13	5.01E+05	4.45E+05	8.42E+05	3.69E+05	2.93E+05	4.07E+05
^{152}Eu	7.65E-03	2.07E-13	8.90E+04	7.10E+04	6.40E+05	4.10E+04	8.00E+05	3.00E+04
^{154}Eu	9.08E-03	2.45E-13	3.80E+04	3.70E+04	2.90E+04	1.70E+04	2.70E+05	2.00E+04
^{155}Eu	7.59E-04	2.05E-14	1.00E+04	8.40E+03	6.30E+04	0.00E+00	7.00E+04	0.00E+00
Total beta (Ci/Kg)			**1.08E-01**	**2.02E-01**	**1.07E-01**	**4.67E-02**	**8.61E-02**	**6.19E-01**
^{233}U	2.86E-02	7.72E-13	1.60E+03	2.80E+03	1.00E+04	1.00E+03	5.20E+03	5.20E+03
^{238}Pu	3.26E-02	8.81E-13	3.80E+03	7.80E+03	5.40E+03	2.40E+03	3.00E+03	1.30E+04
^{239}Pu	3.02E-02	8.17E-13	1.90E+03	3.40E+03	1.30E+03	6.70E+02	8.30E+02	3.40E+03
^{240}Pu	3.06E-02	8.26E-13	8.70E+02	1.80E+03	8.90E+02	3.70E+02	6.00E+02	2.20E+03
^{241}Am	3.28E-02	8.87E-13	3.90E+03	9.30E+03	3.90E+03	2.80E+03	4.60E+03	1.40E+04
^{241}Pu Beta	3.20E-05	8.65E-16	1.40E+04	2.60E+04	1.50E+04	6.50E+03	1.20E+04	2.40E+04
^{244}Cm	3.44E-02	9.29E-13	2.20E+04	5.80E+04	2.80E+04	1.70E+04	2.50E+04	1.10E+05
Total alpha (Ci/Kg)			**1.30E-03**	**2.95E-03**	**1.74E-03**	**8.31E-04**	**1.38E-03**	**4.64E-03**
Total beta in 55 gal. drum (Ci):			30.80	57.05	30.61	14.01	24.55	185.61
Total alpha in 55 gal. drum (Ci):			0.37	0.83	0.50	0.25	0.39	1.39

Table 7.7 Distribution of Decay Heat in MVST Sludge

Isotope	"Q" value (W/Ci)	"Q" value (W/Bq)	Tank W-24 (W/Kg)	Tank W-25 (W/Kg)	Tank W-26 (W/Kg)	Tank W-27 (W/Kg)	Tank W-28 (W/Kg)	Tank W-31 (W/Kg)
^{60}Co	1.54E-02	4.16E-13	1.17E-05	1.04E-05	2.42E-05	5.00E-06	1.75E-05	9.16E-06
^{90}Sr	1.16E-03	3.14E-14	4.39E-05	1.00E-04	2.23E-05	1.41E-05	2.19E-05	3.45E-04
^{90}Y	5.54E-03	1.50E-13	2.10E-04	4.79E-04	1.06E-04	6.74E-05	1.05E-04	1.65E-03
^{137}Cs	1.01E-03	2.73E-14	1.45E-05	1.28E-05	2.43E-05	1.06E-05	8.46E-06	1.17E-05
^{137m}Ba	3.94E-03	1.06E-13	5.34E-05	4.73E-05	8.97E-05	3.93E-05	3.12E-05	4.33E-05
^{152}Eu	7.65E-03	2.07E-13	1.84E-05	1.47E-05	1.32E-04	8.47E-06	1.65E-04	6.20E-06
^{154}Eu	9.08E-03	2.45E-13	9.33E-06	9.08E-06	7.12E-06	4.17E-06	6.63E-05	4.91E-06
^{155}Eu	7.59E-04	2.05E-14	2.05E-07	1.72E-07	1.29E-06	0.00E+00	1.44E-06	0.00E+00
^{233}U	2.86E-02	7.72E-13	1.24E-06	2.16E-06	7.72E-06	7.72E-07	4.02E-06	4.02E-06
^{238}Pu	3.26E-02	8.81E-13	3.35E-06	6.87E-06	4.76E-06	2.11E-06	2.64E-06	1.15E-05
^{239}Pu	3.02E-02	8.17E-13	1.55E-06	2.78E-06	1.06E-06	5.48E-07	6.78E-07	2.78E-06
^{240}Pu	3.06E-02	8.26E-13	7.19E-07	1.49E-06	7.35E-07	3.06E-07	4.96E-07	1.82E-06
^{241}Am	3.28E-02	8.87E-13	3.46E-06	8.25E-06	3.46E-06	2.48E-06	4.08E-06	1.24E-05
^{241}Pu	3.20E-05	8.65E-16	1.21E-08	2.25E-08	1.30E-08	5.62E-09	1.04E-08	2.08E-08
^{244}Cm	3.44E-02	9.29E-13	2.04E-05	5.39E-05	2.60E-05	1.58E-05	2.32E-05	1.02E-04
Total (W/Kg)			**3.92E-04**	**7.49E-04**	**4.51E-04**	**1.71E-04**	**4.52E-04**	**2.20E-03**
Density (Kg/L):			1.37	1.36	1.38	1.44	1.37	1.44
Total in 55 gal drum (Kg):			285	283	287	300	285	300
Total in 55 gal drum (Watt):			**0.112**	**0.212**	**0.130**	**0.051**	**0.129**	**0.660**
Percent of WAC (300Watts) per 3 drums			**0.1%**	**0.2%**	**0.1%**	**0.1%**	**0.1%**	**0.7%**

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