METHODOLOGY FOR DETERMINATION OF RADIOASSAY PROPERTIES FOR RH-TRU WASTE

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ABSTRACT

The Battelle Columbus Laboratories Decommissioning Project (BCLDP) decontamination and decommissioning activities will generate approximately 25 cubic meters of remote-handled transuranic (RH-TRU) wastes, which are planned for ultimate disposal at the Waste Isolation Pilot Plant (WIPP). For WIPP disposal, the isotopic inventory of the waste must be characterized to demonstrate compliance with applicable limits. The BCLDP has developed a methodology that combines limited assay, knowledge of the processes generating the waste, and a dose/weight-to-curie evaluation to determine the isotopic characterization of the waste.

The BCLDP methodology determines the isotopic content for an identified TRU waste stream by a combination of (1) representative waste stream sample analyses, (2) application of Oak Ridge Isotope Generation and Depletion (ORIGEN2) code values for isotopes expected in spent nuclear fuel but not represented by the sample analyses, and (3) assessment of cesium (Cs)-137 content of a payload container based on external radiation field measurements and calculation of TRU isotopic content using a ratio of radionuclides based on known Cs-137 content. The measured isotopic distribution (gamma emitters) of a waste stream is combined with values determined through the conservative application of the ORIGEN2 to account for all isotopes. External dose rates for 1 millicurie of an isotopic mix are modeled for different packaging configurations. Field sort charts permit determining the quantity of radionuclides in each waste package from measured weights and dose rates. The inventory of Cs-137 is determined for each drum based on external radiation field measurement using a high-purity germanium detector. Cs-137 is used as an indicator of TRU content of a drum based on the ratio of radionuclides. The methodology determinations are verified on an approved, periodic basis by gamma and/or alpha spectroscopy. The methodology includes the conservative estimation of measurement errors and assumptions used to determine a total uncertainty that is bounding for the methodology.

This methodology is consistent with worker safety principles and is optimal for the BCLDP, a small quantity site with limited waste and resources. In addition, given the small fraction of RH-TRU wastes to be disposed of at the WIPP, and its insignificant impact on the long-term performance of the repository, the use of sophisticated assay techniques for RH-TRU waste is neither warranted nor necessary. The BCLDP methodology is a simple system that meets the intent of the governing regulations for the WIPP.

BACKGROUND AND PROBLEM STATEMENT

The Battelle Columbus Laboratories Decommissioning Project (BCLDP) decontamination and decommissioning activities will generate approximately 25 cubic meters of transuranic (TRU) wastes, which is planned for ultimate disposal at the Waste Isolation Pilot Plant (WIPP). The major fraction of the total waste to be generated by the BCLDP is remote-handled (RH) TRU waste. A small amount of contact-handled (CH) TRU waste also may be generated. The

BCLDP TRU Waste Certification Program, which is under development, addresses the characterization and certification of both CH- and RH-TRU waste.

If CH-TRU waste is identified, the BCLDP TRU Waste Certification Program plans to subcontract mobile vendors to perform CH-TRU waste radiological characterization. Radioassay of CH-TRU waste containers will be conducted by mobile vendors participating in the Performance Demonstration Program as described in Appendix A of the WIPP Waste Acceptance Criteria (WIPP WAC) (1). The mobile vendors will be responsible for complying with the requirements pertinent to quality assurance objectives; method requirements; quality control; equipment testing, inspection, maintenance, and calibration; and data management, as specified in Appendix A of the WIPP WAC (1).

For the majority of RH-TRU waste, the BCLDP TRU Waste Certification Program proposes to determine its radionuclide content according to the methodology described in DD-98-04, Waste Characterization, Classification, and Shipping Support Technical Basis Document (2). The RH-TRU isotopic content is determined by a combination of knowledge of the processes generating the waste, representative waste stream sample analyses, application of Oak Ridge Isotope Generation and Depletion (ORIGEN2) code values for expected isotopes, and external radiation field measurements. Based on knowledge of the processes documented as "acceptable knowledge" (AK), the contamination of the Controlled Access Area (CAA) of Battelle's West Jefferson North (JN) Building JN-1 has been determined to be representative of contamination in the building as a whole (3). The CAA supported operations conducted in the Building JN-1 High Level, Low Level, Mechanical Test, and Alpha-Gamma Cells, and the Charpy Room. During Building JN-1 operations, the CAA was in constant use and was repeatedly contaminated by numerous projects conducted in all Building JN-1 hot cells. Although the CAA has been cleaned many times, the floors and other surfaces remain contaminated with radionuclides from these projects. As such, smear samples taken from the CAA are considered to be representative waste stream samples and are used as AK in identifying and quantifying the isotopes present in radioactive waste generated in Building JN-1.

STATUS OF WIPP RADIOLOGICAL CHARACTERIZATION REQUIREMENTS FOR RH-TRU WASTES

The radionuclide inventory for each RH-TRU waste container must be quantified to determine compliance with limits for transportation to and disposal of RH-TRU waste at the WIPP. For RH-TRU waste, Table I summarizes the compliance parameters for which radiological characterization is required and the key regulations governing its disposal at the WIPP.

Table I. Radiological Compliance Parameters and Regulations Governing WIPP RH-TRU Waste Disposal

Issue	**Regulatory Agency**	**Radiological Compliance Parameters**	**RH-TRU Waste Regulatory Documents (Reference)**
Transportation	• NRC • DOT	• FGE • Decay heat • Pu content[1] • Description of radionuclides comprising 95% of the radiological hazard	• 72-B Cask Certificate of Compliance issued in 2000 (4) • Amendment to CNS 10-160B Cask SAR under review (5)
RCRA Permit for Mixed Wastes	NMED	Not applicable	Permit modification under development
Disposal of TRU Waste in Repository	EPA-Office of Radiation and Indoor Air	Quantity of Am-241, Pu-238, Pu-239, Pu-240, Pu-242, U-233, U-234, U-238, Sr-90, and Cs-137	Certification decision issued in 1998 (6)
Operations and Safety	DOE-Carlsbad Field Office	• FGE • Decay heat • TRU alpha activity concentration • Pu-239 equivalent activity concentration • Radionuclide activity	RH-WAC to be finalized

[1] Limit on plutonium (Pu) content specified for RH-TRU waste transportation in the CNS 10-160B Cask only (5).

The BCLDP TRU Waste Certification Program baseline approach for transporting RH-TRU waste to the WIPP is the use of the 72-B Cask, a Type B packaging certified by the U.S. Nuclear Regulatory Commission (NRC). Shipment in the 72-B Cask requires quantifying the following properties for which radionuclide composition must be known (7):

- Fissile gram equivalent (FGE)
- Decay heat.

The authorized payload container for the 72-B Cask is the RH canister, which may be directly loaded or may overpack three 55-gallon drums. The capacity of the 72-B Cask is limited to one RH canister (7).

To ensure the timely shipment of RH-TRU waste off-site, the BCLDP also is investigating the potential use of alternative packagings including the CNS 10-160B Cask. An application to include BCLDP RH-TRU waste as authorized payload for the CNS 10-160B Cask is currently under review by the NRC. The application to amend the CNS 10-160B Cask Safety Analysis Report (SAR) (5) proposes limits that require quantifying the following radiological properties:

- FGE
- Decay heat
- Plutonium content.

The authorized payload container is the 55-gallon drum, and the CNS 10-160B Cask can transport ten drums (5).

The RH-TRU waste generated by the BCLDP is destined for permanent disposal at the WIPP. Currently, WIPP waste acceptance criteria is defined only for CH-TRU waste (1). The U.S. Department of Energy (DOE)-Carlsbad Field Office has issued a draft version of the anticipated RH-Waste Acceptance Criteria (RH-WAC) (8). The draft RH-WAC compiles 72-B Cask transportation requirements and anticipated RH-TRU waste characterization and certification requirements for WIPP disposal. The final Resource Conservation and Recovery Act (RCRA) characterization requirements for RH-TRU waste disposal at the WIPP have not yet been approved by the New Mexico Environment Department (NMED) as a modification to the existing WIPP Hazardous Waste Facility Permit. Although the RH-WAC cannot be finalized prior to approval of an RH-Waste Analysis Plan (RH-WAP) by the NMED, the draft RH-WAC specifying radiological characterization requirements, which are not regulated by RCRA, may be used to evaluate the BCLDP proposed methodology for determining radioassay properties. The draft RH-WAC requires quantifying the following properties for which radionuclide composition must be known (8):

- FGE
- Decay heat
- TRU alpha activity concentration
- Pu-239 equivalent activity concentration
- Radionuclide activity
- Ten radionuclides for the purpose of tracking repository inventory curie content (i.e., Am-241, Pu-238, Pu-239, Pu-240, Pu-242, U-233, U-234, U-238, Sr-90, and Cs-137)
- Radionuclides comprising at least 95 percent of the radiological hazard based on A_2 values.

The BCLDP approach for RH-TRU waste characterization is consistent with the draft RH-WAC requirements, which allow the use of AK to determine the required radiological properties.

STATUS OF RADIOASSAY TECHNOLOGY FOR RH-TRU WASTE

Radioassay methods are routinely used to identify and quantify radionuclides in CH-TRU waste. Technologies for determining radioassay properties for RH-TRU waste are being developed. The objective of radioassay performed on TRU waste is to obtain radionuclide composition information to ensure compliance with transportation and waste acceptance criteria related to radiological properties. Radioassay methodologies include both nondestructive (e.g., passive-active neutron counter) and destructive assay (radiochemistry).

Demonstration of nondestructive assay (NDA) of actual RH-TRU waste has been limited. As such, NDA proficiency for RH waste streams is not well known and technology development efforts for RH-TRU waste NDA are in early stages. On the other hand, proficiency of destructive assay on RH-TRU waste is well known and not significantly different from CH-TRU waste. Destructive assay methods are fairly mature but have not been standardized for RH-TRU waste characterization. A significant limitation of the destructive assay of RH-TRU waste is sampling (9).

For much of the RH-TRU waste in the DOE complex, the history, mission and waste management practices of the sites generating the waste are well known. In addition, detailed originating process information pertaining to the physical, chemical, and radionuclide composition of the waste is known. In such cases where the waste-generating processes, including radionuclide composition of the waste, are well documented, assay is not needed. Compliance with transportation and waste acceptance criteria related to radiological properties is ensured based on this process knowledge. Therefore, it has been recommended that knowledge of the chemical and radionuclide content of RH-TRU waste play a larger role in radiological characterization than in current CH-TRU waste activities (e.g., material accountability and control or computations of fuel burnup and reactor fission-product inventory provide comprehensive process knowledge for RH-TRU waste originating from within hot cells). As necessary, process knowledge of radiological characterization may be supplemented by additional information (e.g., as obtained through the use of computer models or limited assay).

The BCLDP is an example of a site for which detailed process knowledge of the RH-TRU waste is known. Building JN-1 operations began in 1955 to support a variety of studies relating to the radiation performance of materials. Experiments in Building JN-1 were largely dedicated to research supporting the DOE (including predecessor agencies) and other government agencies. The research consisted primarily of reactor fuel studies that evaluated materials such as uranium, thorium, and plutonium alloys and compounds in pellet, dispersion, and ceramic form. Control rod material studies included rare-earth absorbers such as europium titanate dispersions in stainless steel. Structural and cladding material studies evaluated stainless steels, zirconium, Zircalloy, nickel alloys, refractory metals, and pressure vessel steels. Building JN-1 primarily supported experiments on small-scale irradiation capsules during the first five years of operations and over the years developed the capacity to examine complete reactor fuel assemblies. The general flow of test materials through the various hot cells and supporting areas of Building JN-1 has been documented. For the BCLDP TRU waste historically generated in Building JN-1, waste management AK information is formally documented in the "Building JN-1 Hot Cell Laboratory Acceptable Knowledge Document" (3).

According to design principles, radioassay methods for RH-TRU waste must (10):

- Provide the data needed
- Be practical for the generator site to apply
- Be consistent with worker safety principles.

Because the aim of radioassay methodology is to collect data for compliance demonstration purposes, a radioassay measurement does not necessarily need to be precise provided that the uncertainty of the method is estimated and conservatively added to the measurement. The resulting value may then be evaluated for compliance with the applicable limit. If the RH-TRU waste is well below the limit, the measurement accuracy has less influence on the compliance determination. In addition, elements that affect relative attenuation in the assay of CH-TRU waste (e.g., waste matrix, density, and location of activity sources within the waste container) have less impact on the overall methodology uncertainty for RH-TRU waste.

This logic provides sites with "the flexibility to weigh the cost versus the benefit of developing higher precision and accuracy analytical methods and optimize for their particular situation….[S]ites may find it more efficient to use less expensive, less precise measurement methods…" (10). The BCLDP TRU Waste Certification Program methodology for determining radioassay properties for RH-TRU waste meets the above design principles and has been specifically designed for implementation by the BCLDP. For example, the use of sophisticated radioassay techniques is not warranted for determining of the fissile content of the BCLDP waste as <50 grams of total fissile material are present on-site (11).

BCLDP PROPOSED METHODOLOGY

The methodology proposed for the radiological characterization of BCLDP RH-TRU waste has been successfully used to characterize low-level waste (LLW) also generated by the BCLDP. The method ensures that the radioactive materials in Building JN-1 are properly sorted as LLW or TRU waste. This methodology is applied only as preliminary characterization for the segregation of CH-TRU waste. As stated earlier, the radioassay properties of CH-TRU waste are determined by assay performed by subcontracted mobile vendors in accordance with the requirements of Appendix A of the WIPP WAC. The LLW and RH-TRU waste radionuclide content are determined as described in DD-98-04, "Waste Characterization, Classification, and Shipping Support Technical Basis Document," (2) and summarized in Figure 1.

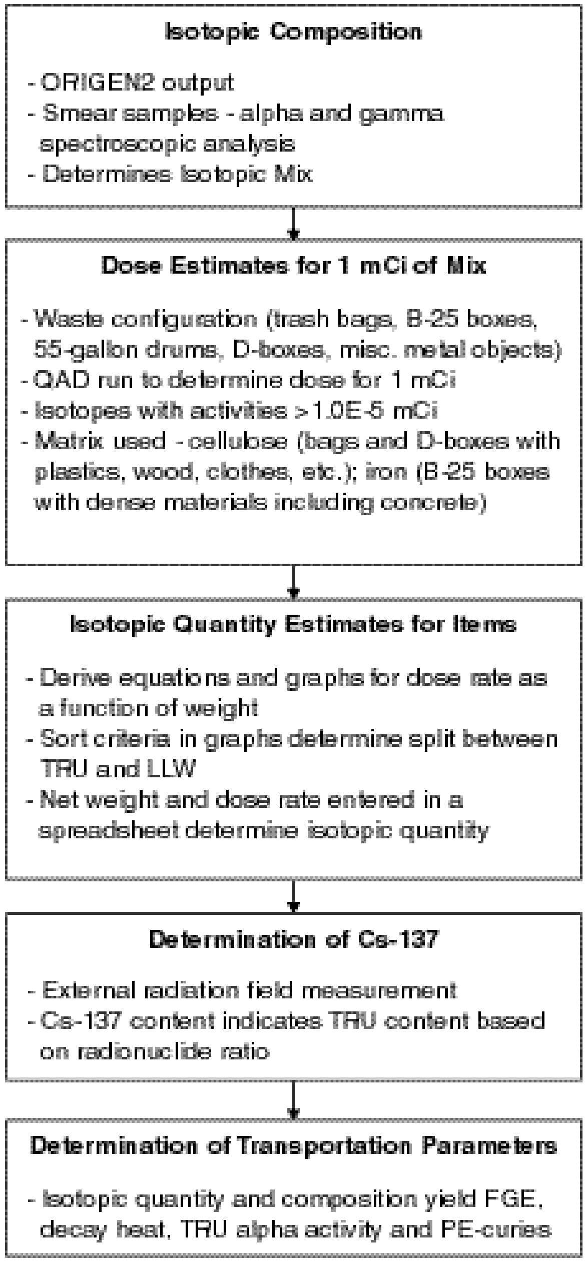

Figure 1

BCLDP Methodology for Determination of Radioassay Properties for RH-TRU Waste

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Under the methodology, the isotopic content for an identified TRU waste stream is determined by a combination of (1) representative waste stream sample analyses, (2) conservative application of ORIGEN2 code values for isotopes expected, but not represented by the sample analyses, and (3) assessment of Cs-137 content of a payload container based on external radiation field measurements and calculation of TRU isotopic content using a ratio of radionuclides based on known Cs-137 content. The determinations are verified on an approved, periodic basis by sample submission to the BCLDP Radioanalytical Laboratory for gamma and/or alpha spectroscopy. The results of the implementation of the DD-98-04 methodology provide the data inputs to the spreadsheets used by the TRU Waste Certification Official to determine compliance with the limits on the transportation and disposal parameters required by the RH-WAC for RH-TRU waste based on the inputs of payload container dose rate, weight, and source.

Use of ORIGEN2 for Determination of Isotopic Mix

Since the gamma rays emitted by radionuclides can be readily detected and quantified by common measurement techniques (i.e., as a dose rate) emitted gamma energies are used to model the quantity of isotopes present in a standard waste stream. Verifying samples are analyzed for both gamma and alpha emitters. Because isotopes other than gamma emitters are known to be present, laboratory measurements of the isotopic distribution are combined with a computer-generated distribution of account for waste site required isotopes, e.g., per U.S. Department of Transportation (DOT) requirements. The measured isotopic distribution is based on laboratory analysis (alpha and gamma spectroscopy) of air, smear, and material samples taken from the accessible work areas of Building JN-1. Using the measured distribution as a base, the remaining isotopes are scaled according to the distribution generated by the ORIGEN2 computer code, which models the production and decay of fission and activation products of commercial nuclear power plant fuel. Commercial fuel best characterizes the overwhelming majority of the isotopes present, by isotope and relative ratio.

The "JN standard isotopic mixture" used in the model is representative of the composition of the majority of radioactive waste generated in all areas of the BCL facility, except the pool. Waste from the pool is characterized separately based on sample results. In addition to the pool waste, other waste streams may be encountered that do not match the JN standard isotopic mix. In such cases, the newly characterized waste will be characterized based on specific analytical results (alpha and gamma spectroscopy).

Characterization of the JN standard isotopic distribution depended upon whether available data existed to permit estimation of the normalized activity ratio (to Cs-137 activity) for the isotopes of interest. Where sufficient data were available, a lognormal fit was used. Where insufficient data were available, the results of a series of ORIGEN2 software analyses were employed.

For Am-241, Cm-244, Co-60, Cs-134, Eu-154, Np-237, Pu-238, Pu-239, Pu-240, Sb –125, Sr-90, U-234, and U-238, as many as 69 samples from the anticipated waste stream were available. A two-parameter (μ, σ) lognormal distribution was fitted to these data. The mean parameter (μ) estimated for each studied isotope represented its assumed normalized activity ratio (to Cs-137) in the standard isotopic distribution. The estimated spread parameter (σ) was used in considering the total uncertainty associated with the waste characterization.

For the isotopes of interest, the computer code ORIGEN2 was used to estimate their normalized activity ratio (to Cs-137). Specifically, values were assumed for enrichment, burn-up, and decay consistent with the processes used to generate the waste stream being classified. These values were then applied as parameters within the ORIGEN2 software, producing estimates of the activities of the various isotopes of interest.

Center, low, and high values for enrichment, burn up, and decay parameters were used to represent the central tendency and distribution (i.e., practical range) of the potential waste streams. The decay time was deliberately underestimated to make the resultant values conservative.

Twenty-seven iterations of ORIGEN2 software code would be required to consider each combination of these three values for each of three parameters (i.e., $3^3 = 27$). Additional code runs would be necessary, moreover, to provide some measure of the uncertainty associated with the application of ORIGEN2 in estimating the normalized activity ratios of the remaining isotopes of interest. Latin Hypercube sampling is an alternative approach, allowing for effective integration of computer code but with fewer runs. To apply these values in the context of a Latin Hypercube design, an assumed distribution is required for each parameter considered in the design. Because the low and high values for each parameter were not symmetric in relation to the center value, a skewed distribution was selected. The lognormal represents a skewed distribution that can be readily applied without additional mathematical complication. The log-transformed center value was assumed to represent the distribution's log mean, and its log standard deviation was derived by averaging the deviations of the log-transformed low and high values from the log mean. Specifically, the average deviation was assumed to represent 1.645 (i.e., the 0.95 quantile of a standard normal distribution) times the log-standard deviation. Doing so is equivalent to assuming the low and high values represent, on average, a range from the 5^{th} to the 95^{th} percentile of the distribution.

In using the ORIGEN2 software code to characterize the normalized activity ratios for the isotopes without available data, the Latin Hypercube employed in the DD-98-04 methodology assumed that values for enrichment, burn up, and decay combined with software values represent a "black box" estimation of normalized activity ratios. Using this approach, a series of replicate designs is applied. The mean result from each replicate design is considered when estimating the mean and variance in normalized activity.

Four replicates of a five-sample Latin Hypercube design were developed thereby providing 20 analysis runs. The distribution of each parameter was divided into five partitions of equal probability. Latin Hypercube sampling, then, ensures that a random value of each partition is included in each of the five replicated designs, while minimizing the total number of required analysis runs.

The mean result across the 20 ORIGEN2 runs (or equivalently, the mean of the mean results determined for the four replicated designs) estimated for each studied isotope represents its assumed normalized activity ratio (to Cs-137) in the standard isotopic distribution. Though ORIGEN2 reports activities for all the isotopes of interest, only the results for those isotopes without sufficient available sample data are retained. The ORIGEN2 results and those based on available data are comparable. The positive correlation between the 69 sample results and the ORIGEN2 results support the characterization of the BCLDP waste as debris from light water reactor fuel. The estimated variance in mean result across the four replicate designs—a measure of the uncertainty associated with using the ORIGEN2 software to characterize isotope activity—is used in considering the total uncertainty associated with the waste characterization.

Use of QAD in Modeling of Waste Matrices and Packaging Configurations

A given quantity of the JN standard isotopic mixture is used as the radioactive material source with the QAD computer shielding code to generate external gamma ray interaction rates for various package and form weights. These interaction rates are used to generate interaction rates-to-weight conversion equations for each package and waste form. The equations are incorporated into spreadsheets so that activity content, in millicuries, for individual packages and waste forms can be calculated. Spreadsheets also are used to calculate TRU interaction rate levels, and plots of these values as a function of net container weight are provided for each container type to simplify field sorting and packaging.

Required QAD inputs include source and package dimensions, including any shielding materials, and quantities of individual isotopes that make up the source (i.e., JN standard isotopic mixture). QAD calculations are performed for a range of representative weights for each package. Specific package models include field sort waste bag, metal case, IP-2 147-cubic-feet box, standard D-box, 55-gallon drum, and standard B-25 box models. The final packaging configuration for BCL RH-TRU waste is the 55-gallon drum.

Waste matrices are modeled as either cellulose or iron. The cellulose matrix represents the varied composition of the bag and D-box models, which are composed of plastics, wood, cloth, etc., and are similar to cellulose in their electronic configuration. The iron matrix is used for the B-25 box and 55-gallon drum models, which include a range of more dense materials, including concrete. The choice of waste matrices is conservative as the physical properties of cellulose and iron relative to radiological parameters are well characterized. It is important to note that the representative weights for the 55-gallon drum, for example, correspond to a density much less than the density of iron (7.86 grams per cubic centimeter [g/cc]), on the close order of less than 1.0 g/cc.

As detailed in DD-98-04, estimated uncertainties associated with the container weight, Cs-137 activity based on measurement of decay gammas emanating from the container, and estimation of inventories of other radionuclides and total transuranics based on measured or predicted ratios to Cs-137 activity have been factored into the determination of an upper bounding uncertainty for the methodology.

SUMMARY

Process knowledge of the chemical and radionuclide content of RH-TRU waste is sufficient for the radiological characterization of RH-TRU waste. Where the radionuclide composition of the waste is well known, assay is not needed. As necessary, such process knowledge may be supplemented with limited assay. The accuracy associated with supplemental assay does not need to be exact provided that a conservative estimate of uncertainty is added to the resulting measurement. If the RH-TRU waste is well below the limit, the measurement accuracy has less influence on compliance determinations. In addition, given the small fraction of RH-TRU wastes to be disposed of at the WIPP and its insignificant impact on the long-term performance of the repository, the use of sophisticated assay techniques for RH-TRU wastes is neither warranted nor necessary, as long as conservative compliance determinations can be made.

At a minimum, methodology for the radiological characterization of RH-TRU waste must provide the isotopic composition and radionuclide quantities, must be practical for the generator site to apply, and must be consistent with worker safety principles. The methodology combining AK, ORIGEN2 modeling, and Cs-137 assessments described in DD-98-04 in the determination of radioassay properties for RH-TRU wastes under the BCLDP TRU Waste Certification Program is consistent with these design principles. Although the DD-98-04 methodology is considered preliminary in that the WIPP waste acceptance criteria for RH-TRU waste have not yet been finalized, the logic supporting the methodology is sound. The BCLDP has obtained the preliminary consensus of other researchers who are developing similar RH-TRU waste radioassay methodologies involving the use of AK. The DD-98-04 methodology is optimal for the BCLDP, a small quantity site with limited waste and resources. The BCLDP methodology is a simple system that meets the intent of the governing regulations for the WIPP.

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