Innovative Applications of In Situ Gamma Spectroscopy for Non-destructive Assay of Transuranic Wastes - 9344

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ABSTRACT

Cabrera Services (CABRERA), under contract to National Security Technologies, LLC (NSTec), supported the transuranic (TRU) waste reduction initiative at the Radioactive Waste Management Complex of the Nevada Test Site (NTS). CABRERA developed advanced NDA techniques for oversized boxes (OSB) and drums using *in situ* gamma spectroscopy during several phases of the project. A more thorough characterization method was employed during the planning phase of the project to better understand the TRU content and distribution within each container, while a comprehensive NDA program was designed and implemented during the intrusive phase that guided waste segregation and re-packaging of both TRU and low-level wastes (LLW).

NSTec took receipt of 58 oversized boxes of suspect TRU waste from Lawrence Livermore National Lab (LLNL). TRU waste is defined as greater than 3.7 kiloBecquerels per gram [kBq/g] (100 nanoCuries(nCi)/g) activity from alpha-emitting radionuclides with atomic number greater than 92 having a half-life greater than 20 years. Each box was custom–made to house a variety of suspect TRU wastes resulting from years of weapons program research, development, and testing. Since their arrival at NTS, the boxes have undergone several iterations of non-destructive assay (NDA) in preparation for the comprehensive repackaging effort. NDA has included two rounds of *in situ* gamma spectroscopy and real-time radiography (RTR) scans that were videotaped. Contents have been confirmed to include glove boxes, HEPA filters and their housings, and assorted process equipment and piping. TRU content was determined via directly measuring plutonium-239 (Pu-239), americium-241 (Am-241), and other radionuclides, while adding calculated results for non-measurable nuclides using reliable scaling factors developed from acceptable knowledge (AK).

Advantages of CABRERA's NDA methods included:

- More NDA information is available in the same amount of counting time, allowing NSTec to make more informed repackaging decisions;
- NDA method was able to report discrete 'hot-spots' within each box, providing vital information used to focus intrusive TRU waste recovery during repackaging;
- TRU determinations could be made immediately for individual waste items extracted from each container; and
- Overall uncertainties of the measured or calculated NDA parameters (mean TRU, Fissile-Gram Equivalent, etc.) were reduced.

Client benefits realized were overall improvements in process efficiency due to less processing in unimportant portions of each box, and reduced volume of TRU waste generated, resulting in substantial cost savings and schedule reduction.

INTRODUCTION

NSTec manages operations of NTS for the Department of Energy (DOE). As part of DOE's waste management services, NTS took custody of 58 oversized boxes of suspect TRU waste from Lawrence Livermore National Lab in California. TRU waste is defined as greater than 3.7 kBq/g (100 nCi/g) alpha activity from radionuclides with atomic numbers greater than 92 and half-lives greater than 20 years. Each OSB was custom–made to house a variety of suspect TRU wastes resulting from years of weapons program research, development, and testing, i.e., gloveboxes, piping, test equipment, etc. Since their arrival at NTS Area 5, the boxes have undergone several forms of NDA in preparation for a comprehensive repackaging effort, currently underway and scheduled to be completed winter 2008. This paper will be updated to reflect the completed project prior to final publication. Prior to CABRERA's involvement, NDA on each box included Real Time Radiography (RTR) scans with the results videotaped and preliminary in situ gamma spectroscopy analyses. As of publication, the project was nearing completion, with approximately six weeks of schedule time remaining.

The purpose of the TRU re-packaging project is to appropriately characterize, package, and ship all TRU wastes in accordance with the Waste Isolation Pilot Plants Waste Acceptance Criteria (WIPPWAC). [1] All waste products determined to meet the NTS WAC as either low-level waste (LLW) or mixed LLW (MLLW) are to be packaged in accordance with the NTS WAC and disposed of at the NTS.

Radiological NDA via *in situ* gamma spectroscopy typically includes high-purity germanium (HPGe) detectors that have been characterized to perform mathematical geometry calibrations. A commercially available package for this purpose is Canberra Industries *In Situ* Object Counting System (ISOCS®). ISOCS® uses mathematical models to simulate radioactive sources in various geometries created from standard geometric templates such as boxes, cylinders, and pipes. These templates are then customized with job-specific information gathered either through direct measurement (dimensions, weight, density) or AK so that the modeled configuration in ISOCS® matches the actual package as closely as possible.

Background of LLNL TRU Waste Stream

Information regarding the waste containers from LLNL was catalogued in a thorough acceptable knowledge document. [2] The TRU waste materials stored at NTS were generated at LLNL between October 1972 and November 1989 as part of research and development of nuclear weapons fabrication and materials research. A summary of the generation of these wastes is excerpted from Ref 1:

"*The waste was originally generated in LLNL Building 251 (Heavy Element Facility), Building 332 (Plutonium Facility), and Building 419 (Hazardous Waste Management Facility). Operations generating waste from various R&D activities and rooms in these buildings include diagnostic testing related to underground testing of nuclear devices, research on the behavior of actinide elements, plutonium metallurgy and other chemical and physical testing, fabrication of plutonium parts, and waste processing (including decontamination and liquid waste solidification). The waste described in this report was generated in defense-related activities including defense research and development, such as post-shot analyses and verification, plutonium recovery, property testing, and defense nuclear waste management*" [2]

A list of the primary TRU radionuclides documented in the AK document [2] are summarized in Table I.

Table I. List of LLNL TRU Waste Radionuclides and Isotopic Distribution

Radionuclide	NDA Quantification Approach (Direct / Indirect)	TRU Nuclide?	Indirect Isotopic Mass Distribution[a]
U-233	Direct	No[b]	n/a
Am-241	Direct	Yes	n/a
Np-237	Direct	Yes	n/a
Pu-238	Indirect	Yes	0.02%[c]
Pu-239	Direct	Yes	93.27%[c]
Pu-240	Indirect	Yes	6.26%[c]
Pu-241	Indirect	No	0.40%[c]
Pu-242	Indirect	Yes	0.05%[c]
Cm-242	Direct	Yes	n/a
Cm-243	Direct	Yes	n/a
Cm-244	Direct	No	n/a
Cm-245	Direct	Yes	n/a
Cf-249	Direct	Yes	n/a

[a] See Ref. 1.
[b] U-233 is not considered a TRU nuclide but is included for WIPP WAC compliance for fissile gram equivalent calculations.
[c] Nuclides determined indirectly were determined by applying the ratio of each nuclide to the Pu-239 result.

Summary of Prior NDA Activities on LLNL Wastes

The previous NDA contractor at NTS used standard *in situ* approaches to characterize the OSBs and drums in inventory. A single-point counting geometry was utilized to assay the mean TRU concentration (in Becquerels per gram [Bq/g] or nanoCuries [nCi]/g) of each container. This mean concentration was calculated by collecting HPGe spectra with the detector positioned at the centerline of each long-side of a rectangular box at a distance of 2 feet. A completed box count is the composite average of two side counts. Although this approach can provide accurate results of mean concentrations within a container, it favors the central portion of the box when reporting the TRU concentration and therefore may not accurately report a true composite average. Principally, a single-point approach relies on the following fundamental assumptions being true:

(1) *The center portion of the box, i.e., the portion of the contents within direct-view of the detector, is representative of the entire box contents (most accurate condition); and/or*
(2) *The center portion of the box contains the highest relative activity (most conservative).*

If neither of these assumptions are true, then the results of the NDA may not yield accurate average TRU concentration measurements. CABRERA utilizes approaches and techniques that eliminate or significantly reduce these uncertainties. Details of both are discussed in the sections that follow.

SUMMARY OF PHASE I NDA ACTIVITIES

CABRERA's support of the TRU activities at NTS was accomplished in two phases. The first phase involved two weeks of on-site NDA evaluations of select OSBs and drums using CABRERA's advanced

NDA methods. Fourteen OSBs initially assayed between 2.8 – 7.4 kBq/g (75 – 200 nCi/g) using the single-point approach discussed previously. Instead of this single-node approach, CABRERA sought to assay as much of the active waste volume as possible as a means for more accurately reporting a 'true' composite average TRU concentration.

The key to this NDA approach is the concept of "Model Reciprocity." Each count represents equal parts of the box, so the model being applied is applicable to all counts being performed as well. Each OSB was carefully measured and marked to ensure reciprocity, plus all model dimensions and offsets were checked for quality control prior to application of the model. This method was implemented by splitting up each side of an OSB into equal sized quadrants and applying a portion of the total count time to each. For example, for a total count time of 40 minutes, the detector would be directed at the center of each quadrant for a total of five minutes, assuming 8 total quadrants, or four per side). After each quadrant was complete, five more minutes was added to the detector 'live time' and the count continued. At the end of the 40 minutes, an eight-point composite average count of the box would be complete and ready for analysis. Each individual quadrant count was also saved using incremental quadrant numbers assigned to the filename, i.e., Q1, Q2, etc. Each count cycle was consistent; starting at the northeast quadrant and moving counterclockwise until all four quadrants on a single side were completed. One side of the box was labeled Q1 through Q4, while the other was labeled Q5 through Q8. The vertical layout of the quadrants was based on 'top of waste' as determined by RTR scans. An example of an OSB quadrant layout is provided in Fig. 1.

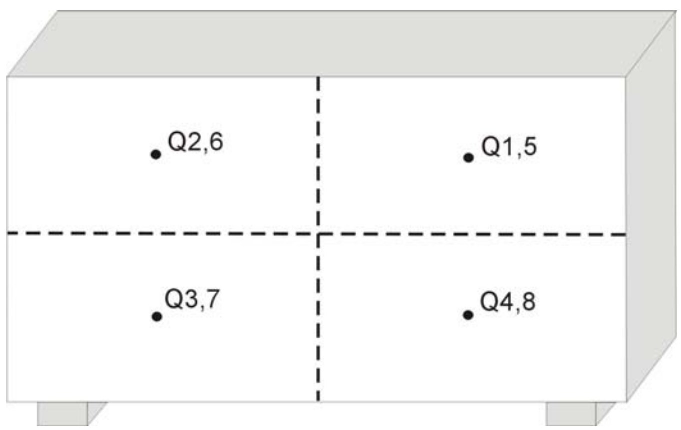

Fig. 1. Diagram of quadrant layout for reciprocity ISOCS modeling of OSBs.

These counts were also performed at a distance between 18-24 inches, depending upon accessibility, as this close distance is appropriate when viewing a reduced portion of the box volume. Each OSB was positioned vertically using a forklift so that the detector centerline could be aligned accurately at each quadrant center. Horizontal movement was performed by mounting the detector in a rolling cart. An example of an OSB (NT 284052) displaying a wide variance in TRU concentration with position is provided in Fig. 2. The levels of measured Pu-239 varied from less than 37 Bq/g (1 nCi/g) to greater than 14.8 kBq/g (400 nCi/g) for the individual quadrants, while the overall average TRU concentration was assayed to be 7.0 kBq/g (188 nCi/g).

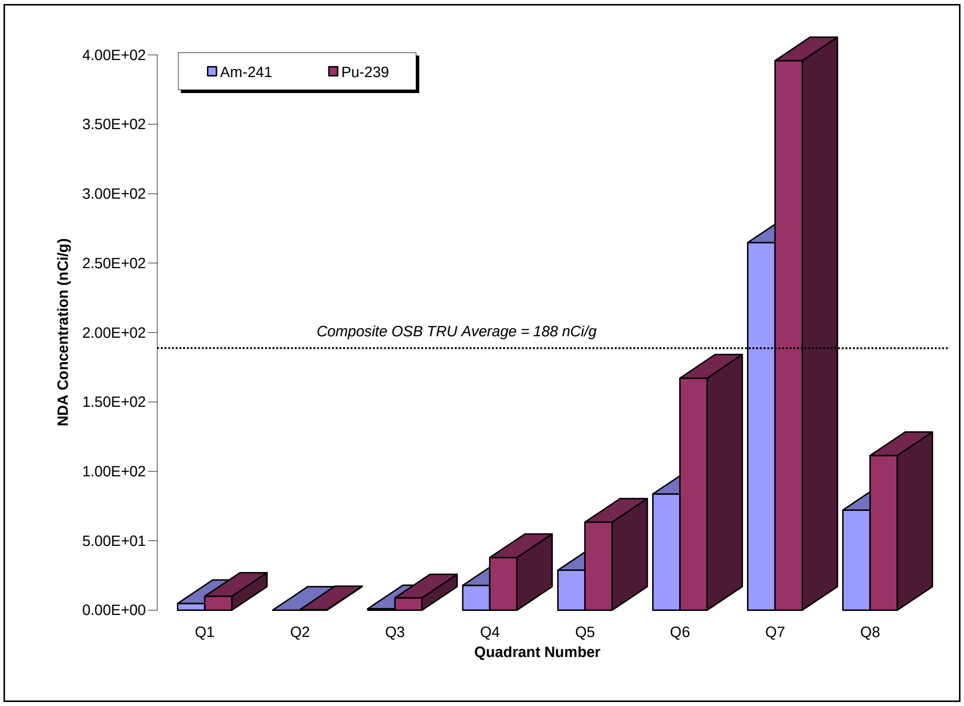

Fig. 2. Plot of Individual Quadrant *In Situ* Gamma Spectroscopy Results For OSB NT284052

The quadrant counts and analysis developed during Phase I (as shown for OSB NT284052) highlighted the advantages of applying advanced NDA techniques to the entire inventory of OSBs during the full-scale TRU repackaging project. Specific advantages realized during the Phase I activities included:

- More NDA information is available in the same amount of counting time, allowing NSTec to make more informed repackaging decisions;
- Ability to identify discrete 'hot-spots' within each container, providing vital information that can be used to focus intrusive TRU waste recovery during repackaging; and
- Reduction in uncertainties of measured or calculated NDA parameters (mean TRU, Fissile-Gram Equivalent, etc.) due to more thorough investigation methods.

PHASE II NDA ACTIVITIES

The success of the Phase I NDA activities led to a follow-up contract with NSTec to support the TRU investigation and repackaging project. CABRERA's NDA methods were integrated into several phases of the Phase II repackaging effort, including:

- Performing a complete NDA characterization of each OSB prior to its being opened. This included a confirmatory count of the average concentration to verify prior NDA results and Generating contoured spatial characterization plots of TRU nuclide gamma fluence on all sides of each OSB to identify hot-spots that could influence intrusive work practices;
- Performing real-time assay counts on extracted waste materials during active repackaging efforts to assist with proper segregation and re-packaging of both TRU and low-level wastes. These counts were referred to as 'Go/No-Go' counts; and
- Performing confirmatory counts on all repackaged wastes containers that exiting the Visual Examination and Repackaging Building (VERB), including completed standard waste boxes

(SWB) slated for WIPP disposal as well as the repackaged OSBs slated for LLW/MLLW disposal at NTS Area 5.

A diagram of the waste container flow through the entire repacking process in provided in Fig. 3, with indicators where NDA processes were integrated. Two independent *in situ* gamma spectroscopy systems were in use during all phases of the TRU repackaging project, one located within the Visual Evaluation and Repackaging Building (VERB) and one located outside the VERB for use in an adjacent drum holding pad.

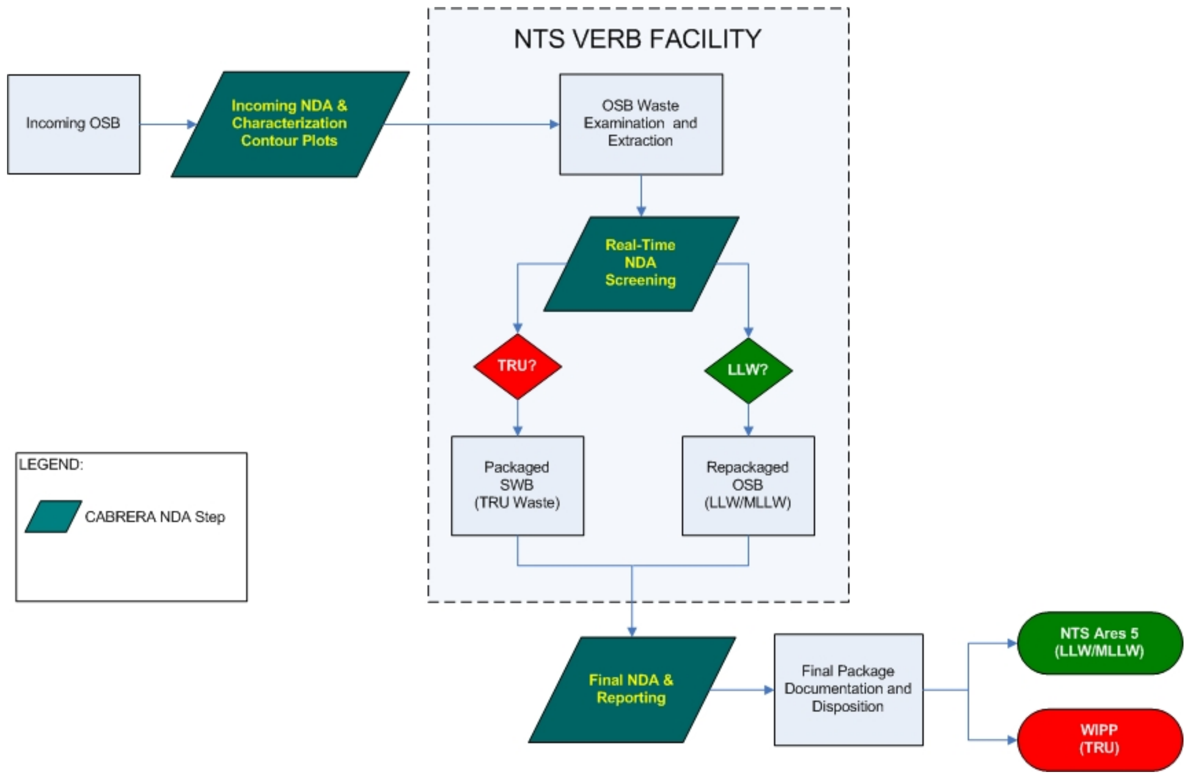

Fig. 3. Work Sequence of Intrusive OSB Repackaging Project with Various NDA Steps Highlighted.

Confirmatory NDA on Incoming OSBs

Each OSB brought to the drum holding pad had a composite average TRU content confirmed via single-point NDA. These counts were performed to verify prior NDA results and planned extraction process before any intrusive work was to begin. These analyses consisted of a long duration count (30 minutes or longer) at a distance of five to seven feet, with the total count time averaged over the two long-sides of each box. Results of each count were forwarded to TRU Operations staff to assist with repackaging decision-making.

Spatial Characterization Plots of OSB Surfaces

The 'multi-node' approach described previously was adapted to produce color contour maps of the gamma fluence exiting all four sides of an incoming OSB. Peak counts from either Am-241 or Pu-239 were logged from up to 16 nodes per side of the box, then gridded and contoured in a geospatial mapping program. The detector and supporting computer/electronics were mounted on a portable, battery-powered cart with a manually-operated scissor lift. This platform allowed for precise locating of the detector at the various node locations up to a height of six feet. The HPGe detector was also outfitted was a 90-degree lead collimator for this application to reduce the active field of view, thus improving spatial resolution of the data.

Data were collected using short count times (30 seconds to 2 minutes) since the target information was net peak counts only. The portability of the system allowed for quick repositioning of the detector at the various node locations, with total data collection time averaging only 15 to 30 minutes per OSB.

The outcome of these counts is a color contour map of the gamma fluence with respect to position that can be used as a supplemental NDA tool to guide repackaging efforts. A set of completed spatial characterization maps for a re-packaged OSB is shown in Fig. 4.

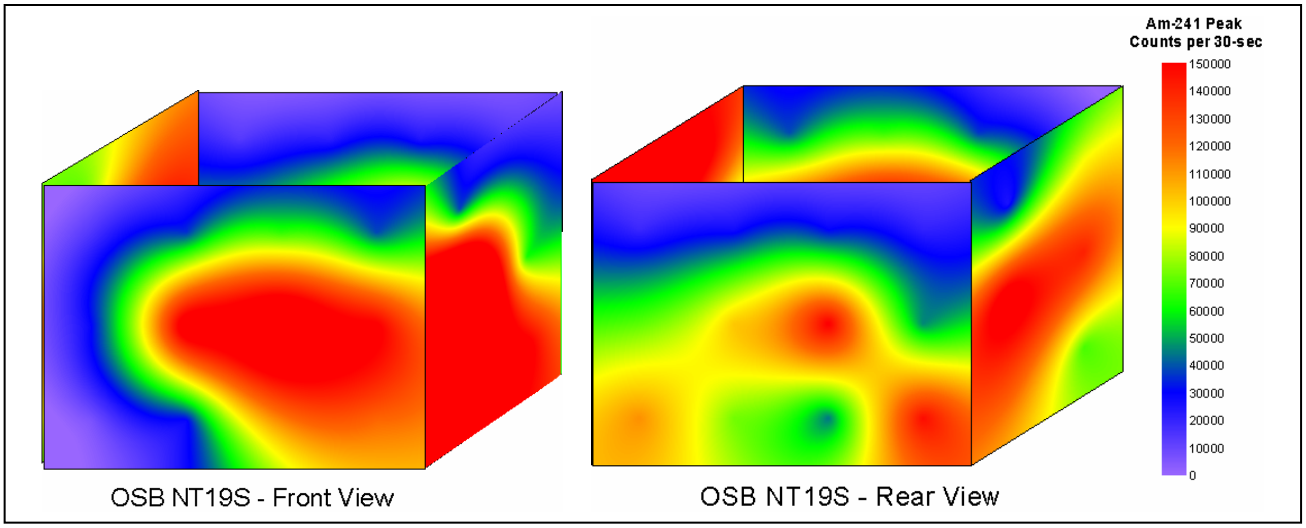

Fig. 4. Color Contour Maps of ^{241}Am Gamma Fluence From Outer Surfaces of OSB NT19S

On two occasions, the surface contour maps, when used in concert with available RTR imagery, were successful in identifying discretized items of concentrated TRU activity within an OSB that were causing the entire composite TRU average concentration to exceed 3.7 kBq/g (100 nCi/g). For these OSBs, precise mining and extraction of the of the offending items resulted in very fast turnaround of a target OSB, saving valuable schedule time and resources in the process.

Examples of these discretized items included loaded HEPA filters, sources, and heavily contaminated pieces of process equipment such as piping. These contour maps came to be known as 'MRI-Plots' by the NTS crew as an indication of their value and utility to the project.

Real-time NDA of Extracted Waste Materials

CABRERA's NDA expertise allowed NSTec to perform live 'Go/No-Go' determinations on select wastes so that the total quantities of TRU waste generated could be minimized. These counts were performed by counting extracted waste items as they were removed and analyzing the results using efficiency models generated 'on the fly' using the ISOCS software. Having this ability in the field offered NSTec the opportunity to realize tremendous cost savings, principally by maximizing the quantities of LLW that could remain at Area 5 for direct disposal.

All repackaging operations were performed in a permanent contamination structure (PermaCon) that was previously constructed inside the VERB. The PermaCon was designed to house posted "High-Contamination Area" (HCA) and "Contamination Area" (CA) to accommodate processing of TRU wastes. Due to the radiological conditions within the PermaCon, CABRERA designed and engineered a wireless *in situ* gamma spectroscopy solution that enabled remote control of the detector and electronics via an adjacent control room located outside the posted radiological boundaries. The HPGe was located within the CA with a direct view into the HCA, where active repackaging efforts were being performed. It was decided during organization review meetings that it was not practical to locate the HPGe within the HCA, as there would be a high probability of cannibalization due to internal contamination. A compromise solution placed the detector within the CA in a location with a direct, unimpeded view of the HCA. NDA shots of suspect TRU items were performed through a roll-up door made of several layers of reinforced plastic sheeting, which served as the primary passageway for waste packages into the HCA. The HPGe detector was allowed to be filled with liquid nitrogen over the CA boundary, such that release surveys by radiation control technicians were not be required prior to each routine re-fill. This also saved the project valuable time and resources.

Assay of suspect waste items was accomplished by having HCA staff remove the item from the OSB, measure and weigh it, then describe its makeup and construction to the ISOCS operator over radio communication. The ISOCS Operator would then initiate a count and begin modeling the object to obtain a calibration file used to estimate its activity concentration. CABRERA staff quickly modeled a variety of extracted waste items including gloveboxes (both intact and various individual parts), a variety of process equipment (filters, pumps, piping, etc), and bags of assorted waste, personal protective equipment (PPE), and foam. As new items were encountered, models were added to the library until a working set of basic models were available for even faster NDA results. Implementation of a previously established model would only require minor adjustment for weight, density, or material prior to its use.

The HCA crew was equipped with handheld alpha, beta, and gamma detectors for monitoring the radiological conditions during active repackaging. The gamma detectors (both dose rate and count rate) were also used to pinpoint the locations of TRU waste items identified during the spatial characterization of the OSBs. In several cases, the crew was able to find small items of relatively high TRU activity that were the primary sources of the entire OSB being considered TRU waste. In these cases, the items were confirmed as TRU via NDA, and the remainder of the OSB contents investigated. If the remainder of the OSB was confirmed as being less than 3.7 kBq/g (100 nCi/g), re-packaging operations for the OSB were discontinued. The OSB would then be reassembled and wrapped for processing as a LLW package. For those OSBs where no specific discretized items of high activity were identified, the 'Go/No-Go' counts were continued until all confirmed TRU waste items were removed.

Real-time assay of removed waste materials was challenging at times due to increased background levels within the relatively small confines of the HCA (less than 100 square meters). Annular lead shields and collimators were installed around the detector to reduce the effects of these backgrounds and also took advantage of angular shot paths away from adjacent wastes to maximize the benefits of the installed shields from directly incident photons.

Catalyzing foam was also considerable challenge during the repackaging operation. It was discovered that many OSBs were filled with this foam during waste loading as either a means of complete encapsulation or as blocking and bracing. All of this foam had to be removed in order to comply with the WIPP waste acceptance criteria, so it was systematically removed from the affected boxes, inspected, and surveyed. The extracted foam was placed in large plastic bags and assayed using a collection of standard models based on size and weight. Bags of foam found to be less than 3.7 kBq/g (100 nCi/g) were characterized as LLW and repackaged into the OSBs, while foam assayed as TRU waste was loaded into SWBs.

Final NDA on All Completed Packages

Confirmatory NDA was performed on all completed SWBs, re-packaged OSBs, and drums that exited the VERB. Each container was temporarily brought to the adjacent holding pad for NDA prior to its being moved to a secure location. The same technique described for the incoming NDA counts was also applied at this stage. This confirmatory NDA served as the final radiological results for all TRU program documentation and reporting. Radioassay Data Packages were generated for each completed package that contained the following information:

- Activity concentrations and uncertainties for each measured or inferred radionuclide;
- Determination whether package was TRU or LLW;
- Pu-239 Fissile Gram Equivalent and Associated Uncertainty;
- Pu-239 Equivalent Mass and Associated Uncertainty (in grams)
- Pu-239 Equivalent Activity (in Curies);
- A copy of the ISOCS model summary; and
- A copy of the gamma spectrum analysis printout.

The Pu-239 Equivalent Mass and Equivalent Activity values were evaluated in accordance with the WIPP Waste Acceptance Criteria. [1] The concept of Pu-239 equivalence lies in the ratio of inhalation dose-conversion factors for Pu-239 and all other TRU nuclides present in a given container. This calculation simplifies the evaluation of the potential long-term dose consequence of incoming payload containers at WIPP by streamlining the nuclide activities to a Pu-239 equivalent value.

RESULTS

The NDA processes described herein contributed to a considerable reduction in TRU waste volume requiring disposal at the WIPP facility, which in turn increased the volume of waste that could stay at NTS Area 5 as either LLW or MLLW. As of November 4, approximately 65% of the total net weight of the incoming OSB wastes (over 26,000 kilograms) has been retained at NTS for disposal at their Area 5 facility. ‘Net weight’ as described here indicates that the tare weight of the container itself was not included in these calculations.

The increased amount of waste determined not to be TRU has had a profound impact on the anticipated volume of SWBs requiring disposal at WIPP. An estimated TRU waste volume savings of 200 cubic meters (m^3), or 120 SWBs, is projected as a result of improved waste characterization, identification, and processing capabilities within the VERB facility at NTS Area 5. These savings were realized in part through the improved NDA capabilities described previously, which allowed NSTec management and staff to focus efforts and better utilize resources to process the OSB wastes most efficiently.

CONCLUSIONS

The advanced NDA techniques described in this paper provided several significant benefits to NSTec during the TRU waste repackaging project. These include:

1. **Significant Increase in Available NDA Information** –NSTec received more information in the same amount of time via innovative solutions customized to their needs. Several of the innovations developed for this project became key components that were used daily.
2. **Reduction of Cost & Schedule** – The additional NDA processes were shown to be cost-effective in that ~66% of the re-packaged waste (by mass) was retained at NTS for LLW disposal. This was achieved through the use of ‘Go/No-Go’ in-process NDA capability as well as the thorough characterization that led to focused TRU item removal. Several OSBs

were repackaged very quickly once it was discovered that the overall TRU levels were being driven by just a few discrete items.

3. **Reduction of Risk** – NDA also contributed to an overall reduction in project risk on several fronts.
   a. *Correct Waste Categorization* – In-process NDA provided a high level of confidence that all segregated wastes were being correctly classified (TRU vs. LLW). Without this capability, it is likely that much more of the re-packaged waste would have been conservatively disposed of as TRU.
   b. *Non-compliance with Waste Acceptance Criteria* - Each and every container brought out of the examination and re-packaging facility (both TRU and LLW) was verified ensure compliance with either the NTS or WIPP WAC. CABRERA's data packages were also used as the official values of record for all packages.

This level of information fostered efficiency gains through improved decision making by NSTec Project Management staff. Relationships between packaged components and their TRU content was more easily recognized up front, which led to cost savings through more efficient use of time and resources, and reduction in TRU waste volume.

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