Low Level and Transuranic Waste Segregation and Low Level Waste Characterization at the 200 Area of the Hanford Site

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ABSTRACT

This paper describes the waste measurement and waste characterization activities carried out by ANTECH Corporation (ANTECH) and CH2M Hill Plateau Remediation Company (CHPRC) at the 200 Area of the HANFORD Site under Contracts No. 22394 and No. 40245 for the US Department of Energy (DOE). These include Low Level Waste (LLW) and Transuranic (TRU) Waste segregation and LLW characterization for both 55-gallon (200-litre) drums with gross weight up to 454 kg and 85-gallon over-pack drums. In order to achieve efficient and effective waste drum segregation and assay, ANTECH deployed an automated Gamma Mobile Assay Laboratory (G-MAL) at the trench face in both 200 Area West and East. The unit consists of a modified 40 foot ISO shipping container with an automatic flow through roller conveyor system with internal drum weigh scale, four measurement and drum rotation positions, and four high efficiency high purity Germanium (HPGe) detectors with both detector and shadow shields. The unit performs multiple far-field measurements and is able to segregate drums at levels well below 100 nCi/g. The system is sufficiently sensitive that drums, which are classified as LLW, are characterized at measurement levels that meet the Environmental Restoration Disposal Facility (ERDF) Waste Acceptance Criteria (WAC). With measurement times of between 20 and 30 minutes the unit can classify and characterize over 40 drums in an 8-hour shift. The system is well characterized with documented calibrations, lower limits of detection (LLD) and total measurement uncertainty. The calibrations are confirmed and verified using nationally traceable standards in keeping with the CHPRC measurement requirements. The performance of the system has been confirmed and validated throughout the measurement process by independent CHPRC personnel using traceable standards. All of the measurement and maintenance work has been conducted during the period under a Quality Assurance Plan (QAP) compliant with the applicable criteria of NQA-1 (2000). This includes not only the calibrations and measurements but also the data analysis activities of the ANTECH Subject Matter Experts (SME) and ANTECH support and maintenance activities as well as the activities of CHPRC staff who recover, transport and load waste drums and disposition measured and characterized drums. The overall processes of drum recovery and analysis are described in the paper. Specific spectral data is presented which illustrates the segregation, sentencing and assay process for different types of drums with different radionuclide profiles. The process of identifying and quantifying a wide range of non-TRU radionuclide isotopes is explained and illustrated with spectral examples. The difficulties associated with the measurement of drums with a high gamma ray background, usually arising from high levels of Cs-137 are considered. These drums, which would normally be declared indeterminate and treated as TRU, are addressed under contract No. 40245 by the deployment of the ANTECH Neutron Mobile Assay Laboratory. This is an Active-Passive neutron assay system housed in a modified ISO shipping container. The unit is designed for the measurement and assay of both drums and crates (including B-25 boxes and SWB containers) and will quantify the content of both plutonium and uranium. The neutron system has been employed to perform further evaluation on indeterminate drums to classify them to either LLW or TRU. The experiences of both gamma

ray and neutron system operation in different conditions are described; as are the issues of throughput, drum handling and system maintenance. All of these are considered in the over-riding context of safe drum handling and safe assay system operation.

INTRODUCTION

Cleanup of the Central Plateau at the Hanford Site, Richland Washington, includes remediation of the Low Level Waste (LLW) burial grounds containing suspect Transuranic (TRU) waste. TRU wastes were retrievably stored in the Low Level Burial Grounds (LLBGs) from 1970 through 1987. CH2M Hill Plateau Remediation Company (CHPRC) is the U.S Department of Energy contractor tasked with the responsibility to retrieve radioactive solid waste from the burial grounds, to assay suspect TRU waste utilizing nondestructive techniques, and to characterize waste as TRU or Low-Level Waste (LLW).

ANTECH Corporation (ANTECH) has provided nondestructive assay (NDA) and waste characterization support to both Fluor and CHPRC in the 200 Area Burial Grounds since 2004. ANTECH have developed an NDA service to classify and segregate suspect TRU from LLW in both 55-gallon and 85-gallon over-pack drums and to characterize LLW for final disposal. The NDA systems are mobile and contained in ISO shipping containers. Although the systems are operated by local staff managed by CHPRC, ANTECH provides an on-site supervising operator, maintenance and calibration support. Data analysis, review and electronic data pack QA report-preparation are carried out by ANTECH subject matter experts (SME), who are based at the ANTECH facility in Westminster Colorado.

The purpose of this NDA system is to classify suspect TRU waste as LLW or TRU as defined by DOE G 435.1-1 (Chapters III and IV). The NDA equipment performs measurements that accurately and reliably process NDA assay results with sufficient confidence to distinguish TRU waste from LLW. Gamma ray assay protocols are primarily used to quantify and classify TRU waste. The ANTECH gamma ray detector configuration is also sufficiently sensitive to characterize LLW at measurement levels that meet the Environmental Restoration Disposal Facility (ERDF) waste acceptance criteria. Recently, the ANTECH assay equipment was enhanced to include active/passive neutron assay to sentence high gamma ray dose drums that would have had an indeterminate gamma ray assay.

The essential deliverable for this project is a final batch data report, with sufficient Quality Assurance documentation to support the findings, of NDA classification as TRU or LLW for each measured waste container.

EQUIPMENT

The ANTECH mobile NDA measurement systems are properly calibrated, verified, and validated and therefore qualified for characterization of TRU/LLW. The gamma mobile assay laboratory (G-MAL) has the capacity to perform 4 simultaneous far-field HPGe drum measurements. The normal drum measurement duration is 20 to 30 minutes. Applying this measurement protocol while employing very conservative logistical consideration enables the measurement of 40 suspect TRU waste drums per measurement day with relative ease. The neutron mobile assay laboratory (N-MAL) is an active/passive counter contained in an ISO shipping container capable of measuring either crates or drums. The G-MAL and N-MAL can be viewed in Fig. 1-3.

The G-MAL is self-contained and designed with physical capabilities to accept, weigh, move, and manipulate 55 or 85-gallon waste drums with gross weights up to 454 kg. The G-MAL, N-MAL and all associated equipment are fully compliant with National Fire Protection Association (NFPA) and National Electric Code (NEC) requirements.

Figure 1. The ANTECH Gamma Mobile Assay Laboratory (G-MAL) located at the trench face in the 200 Area West at the Hanford Site.

ANTECH provides officially documented system calibration, Lower Limit of Detection (LLD), and Total Measurement Uncertainty (TMU) documentation [1] and the NDA system's LLD is defined as a minimum detectable concentration (MDC) of TRU isotopes sufficiently below 100 nCi/g to distinguish TRU from LLW. Calibration confirmation demonstrates that the NDA system is within an acceptable operation range as established by the CHPRC measurement requirements.

Figure 2. Interior of the ANTECH Gamma Mobile Assay Laboratory (G-MAL) with test drums.

Calibration Verification demonstrates that the system accuracy and precision comply with CHPRC measurement requirements.

The measurement protocol is directed through a graphical user interface. Resident software control interfaces with programmable logic controllers in the G-MAL to drive conveyors and measurement platforms for reliable and reproducible performance in routine operations. NQA-1 certified software minimizes the operator interface and reduces the number of repetitive, mundane tasks allowing the skilled operator to focus on higher-level production and operation activities. The N-MAL is loaded using a forklift truck for single drum measurements. ANTECH provides an operational procedure, maintenance procedure, and troubleshooting guide that are utilized by the ANTECH supervising operator and SMEs to ensure proper and efficient operation of the NDA systems.

Figure 3. The ANTECH Neutron Mobile Assay Laboratory (N-MAL) during factory testing.

Throughout the measurement period, instrument performance, with respect to quality assurance and quality control, is displayed through measurement control charts. CHPRC personnel complete independent performance assessments to validate and attest to the operational probity of the NDA systems. The final batch data report is a compilation of measurement-data, quality assurance documentation, and NDA data analysis assembled in an appropriate electronic data file format for transmission to CHPRC personnel. This report is authored, reviewed, and approved by ANTECH personnel who are qualified to CHPRC requirements for NDA measurement review.

MEASUREMENT LOGISTICS AND ORGANIZATION

The NDA systems are deployed in the Low Level Burial Grounds (LLBG) near the location of the buried waste containers. The systems are, however, mobile and can be readily relocated to undertake measurements in an alternative location. Since 2004, the ANTECH assay equipment has been deployed in both the 200 West and 200 East Area waste burial grounds. Prior to entry on-site the CHPRC Radiological Control Organization defines and reviews the adequacy of the ANTECH emergency and operating procedures, current licenses, registrations (if applicable), personnel dosimetry history, and list of authorized users before authorizing the installation of the NDA system. CHPRC staff also managed sources provided by ANTECH.

CHPRC provides electric power and operators for the NDA assay equipment. The CHPRC operators work in cooperation with the ANTECH supervising operator. CHPRC Engineers perform initial and routine safety walk-downs or inspections ensuring NFPA and NEC compliance prior to initiation of NDA work. Measurement support activities, such as drum staging, preparation, delivery, return, and radiological control support are provided and coordinated by CHPRC operations management.

CHPRC personnel are provided access to analysis hardware/software so that reviews and surveillances of assay activities are verified in order to validate that the measurement protocols in place are adequate and effectively support the CHPRC waste measurement mission for NDA.

ANTECH monitors background radiation fluctuations due to container movement to ensure that changing storage configurations and shifting background radiation levels do not adversely affect assay measurements. Upon completion of a measurement batch, which is generally processed on the basis of a measurement day, an assay results report that identifies each measured drum as “TRU” or “LLW” is submitted as a preliminary result. Individual analyses that identify isotopic interferences or measurement configurations that may alter this preliminary characterization are completed by an ANTECH isotopic analysis specialist (SME) and submitted with the final batch data package.

NDA assay work is not performed in areas with radiological contamination. The ANTECH assay labs are located near the burial trench face within the boundaries of a Hazard Category 2 Nuclear Facility in either a radiological buffer area (RBA), radioactive material area (RMA), radiation area (RA), or high radiation area (HRA). The measurement location is a field operation and daily operations are coordinated through first level management and CHPRC Field Work Supervisors. Complete operational authority, from personnel scheduling to hours of operation, are directed by CHPRC. All ANTECH work activities are governed by CHPRC approved Procedures or Work Packages.

ANTECH measurement and maintenance personnel operate to an NQA-1 (2000) compliant Quality Assurance Plan (QAP) that implements, maintains, and documents performance of NDA activities. They are responsible for system operability, maintenance of NDA system calibration and operations safety including the operability of safety interlock systems. ANTECH SMEs and support personnel provide both routine and special maintenance for the NDA systems in compliance with Environmental, Health and Safety requirements and CHPRC site operating procedures.

CHPRC personnel provide all drums for measurement by the NDA systems. Drums that are physically acceptable and have radiological approval for measurement by the NDA system are staged and loaded for measurement in the NDA systems. CHPRC operators perform measurement of the waste drums under the supervision of the ANTECH supervising operator.

DRUM RECOVERY AND MEASUREMENT

Some 6,000 drums were recovered and measured by the G-MAL from February 2004 to August 2009 from the 200 Area West of the Hanford Site under Contract No. 22394 initially under Fluor Hanford and subsequently under CHPRC for the US Department of Energy (DOE). The drums consisted of both 55-gallon drums and 85-gallon over-pack drums. In general the initially recovered drums were in sound condition as they had been place in the trenches in a vertically stacked configuration and were the last drums placed in storage. As the length of time in storage increased the condition of the drums deteriorated. By 2007 nearly all drums were being

over-packed in 85 gallon drums so that drum recovery was generally less difficult than for drums recovered from trenches in the 200 Area East.

In this paper we will concentrate on the most recent drum recovery activities, conducted by CHPRC from the 200 Area East of the Hanford Site. ANTECH NDA activities in association with the drum recovery were conducted under CHPRC contract No. 40245 for the US Department of Energy (DOE). ANTECH deployed both the G-MAL and the N-MAL to the trench face in the RMA in the 200 Area East of the Hanford Site. The measurements were performed on site. Recovered drum configurations were: recovered intact 55-gallon drums, recovered 55-gallon drums that were overpacked with 85-gallon drums, or recovered materials that were placed into new 55-gallon drums. More than 1500 drums, approximately two-dozen boxes, and a few miscellaneous configurations of contaminated soil and orphaned items from breached drums were recovered during the excavation period of January 2011 through September 2011.

Drum Recovery

The burial style and the overburden material used in the 200 East trenches impeded drum recovery and these factors caused drum excavation to become a labor-intensive activity that delayed drum removal. The drums were placed in the trench horizontally and covered with a rocky soil overburden. The heavy excavation equipment could not be employed in the removal of the rocky overburden, near the drums, for fear that the excavator might damage or rupture the waste containers. These difficulties were not anticipated until recovery personnel were deeply into the drum removal process. Previous drum removal experiences had not encountered these unique excavation difficulties, which led to diminished throughput and consequent project delays.

Figure 4. Drums and debris uncovered in a trench of the 200 East Area burial ground. The configuration of the drums necessitates manual excavation and handling.

The excavated drums are recovered into drum bags and removed from the trench either individually or as a group on pallets. A rudimentary triage is performed based upon the drum weight, handling characteristics, and the in situ radiological information reported for each drum. High dose drums, i.e., drums with surface dose readings greater than 35 mrem per hour, are

isolated and scheduled for separate measurement. This segregation is performed in order to reduce personnel exposure and to acknowledge ALARA considerations. High dose drums are measured two at a time in the outside measurement positions in the G-MAL, reducing throughput. If a high dose drum is indeterminate due to interference in measuring the 414 keV gamma ray line from Pu-239, then it is subsequently measured using the N-MAL.

Drums that are slightly distorted, but not breached, or are very heavy are staged in the north drum staging location; however, these drums are physically separated from the normal waste drum population. This segregation is performed to identify drums that needed additional attention or caution when they are moved for measurement. All other bagged drums are then staged in the north drum staging location. No drum segregation is performed for these bagged drums and they are intermingled with the existing (normal) waste drum population for measurement.

Drum Measurement and Analysis

Far-field gamma ray measurements are performed on both 55-gallon and 85-gallon overpack waste drums using the G-MAL. Drum contents are unknown; however, a coherent acceptable knowledge profile has been developed for this drum population. The historical knowledge of waste generation and waste constituents is most useful in bracketing the isotopic search parameters and allows the development of a concise library for routine analyses.

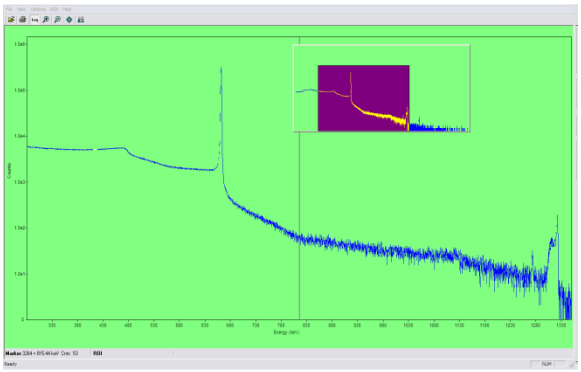
The drum population for contract No. 40245 divides into four generic isotopic populations. There is a small population of drums (approximately 5%) that display an intense signature and associated Compton continuum for Cs-137 where gamma ray signatures below 661 keV are either distorted or obscured. For these samples the identification of primary signature peaks that are less than 661 keV is ineffectual and isotopes with energy signatures below 661 keV are difficult to quantify. These indeterminate drums are subsequently measured using the N-MAL.

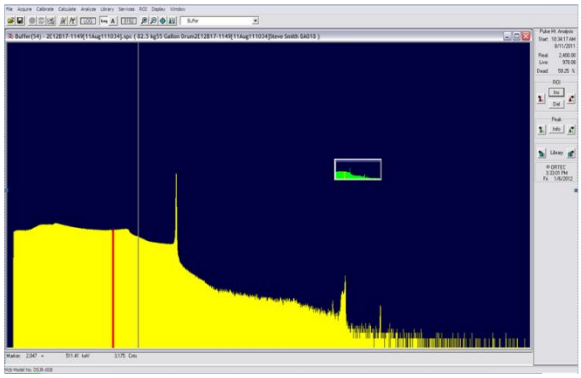
A second generic population consists of samples that contain quantifiable amounts of U-233. Although U-233 is not a TRU radionuclide this population has samples that may lead to a TRU or LLW characterization. A third population consists of samples that have a significant TRU radionuclide signature and are characterized as TRU waste drums. Isotopes other than Pu and Am, that may or may not be TRU radionuclides, are identified in these samples. And the fourth significant waste population has drums that do not have a substantial TRU radionuclide signature and are characterized as LLW. These waste drums may have TRU radionuclides but the TRU waste content is below 100 nCi/g and they are therefore characterized as LLW.

Drums with Cs-137 content that display a distinctive full energy peak at 661 keV and an intense Compton continuum from this energy down are not favorably disposed to gamma ray isotopic analysis for Pu and Am. Isotopic peak signatures in the 50 – 661 keV region are not useful and energy signatures below the 1323 keV sum peak may also be compromised by an overwhelming Cs-137 presence. Because standard isotopic analysis is not useful in calculating the isotopic composition or efficacious in ascertaining the TRU activity for these samples alternative analysis means must be applied.

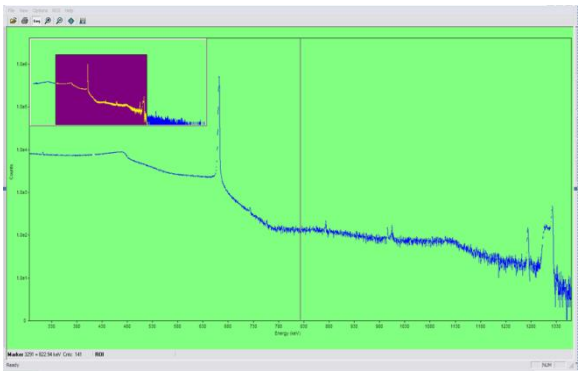
For the specific consideration of this contract passive neutron interrogation is an effective means of characterizing these high Cs-137 samples as TRU or LLW. Using the N-MAL, the neutron measurement determines the fissile component of the sample and acceptable knowledge profiles establish the likely isotope composition for the sample. The acceptable

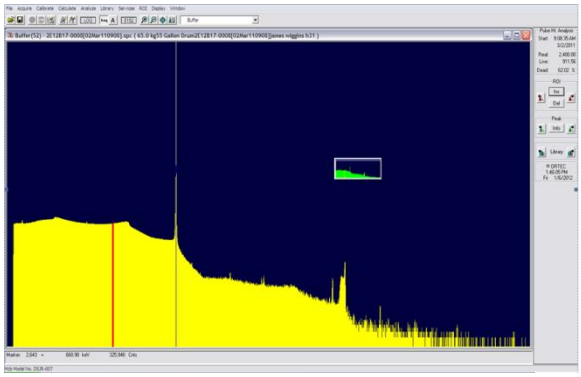
knowledge profile also helps to ascertain if neutron coincidence counting or special algorithms for alpha-n interactions are necessary.





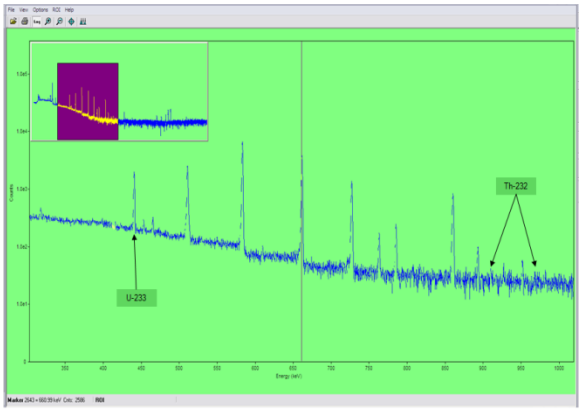
Figures 5a and 5b. Cs-137 indicator @ 661 keV and sum peak @ 1323 keV, no Pu indicated and the neutron measurement reports Pu-240 below detection limit. Lower detection limit relates to 0.045 g total Pu in the sample.

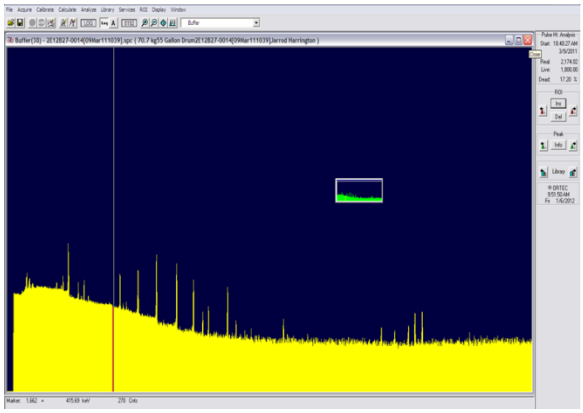




Figures 6a and 6b. Cs-137 indicator @ 661 keV and sum peak @ 1323 keV, no Pu indicated; however, the passive neutron measurement reports 22.27 mg Pu-240 related to 0.167 g total Pu in the sample.

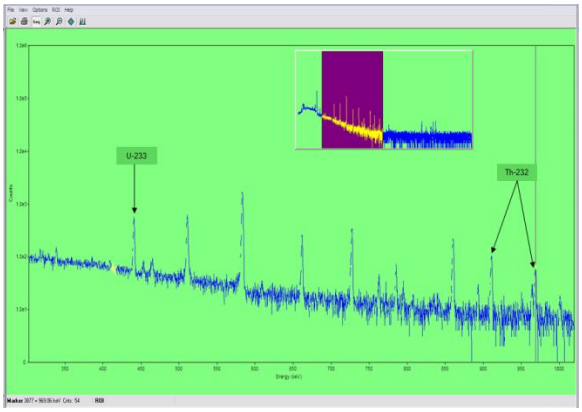
Samples that contain quantifiable amounts of U-233 are relatively unique, but the acceptable

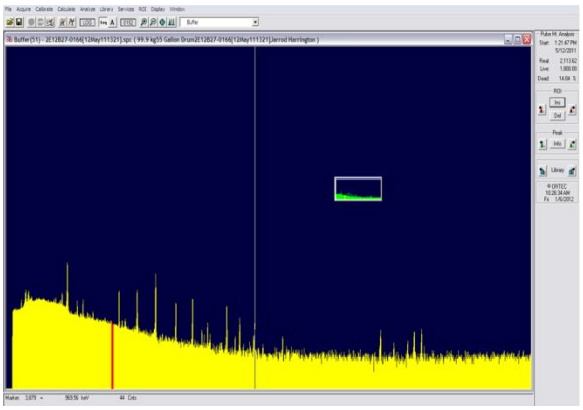




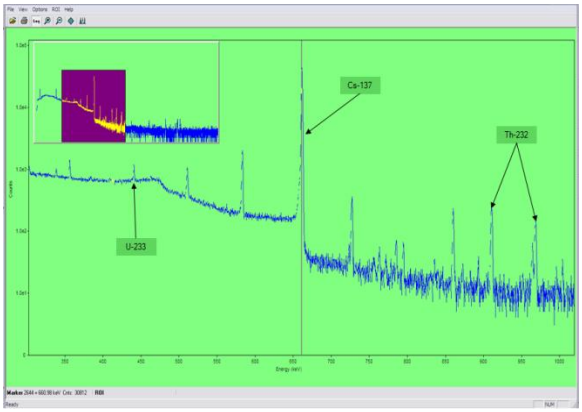
Figures 7a and 7b. U-233 indicator @ 440 keV, no Th-232 indicators @ 969 and 911 keV

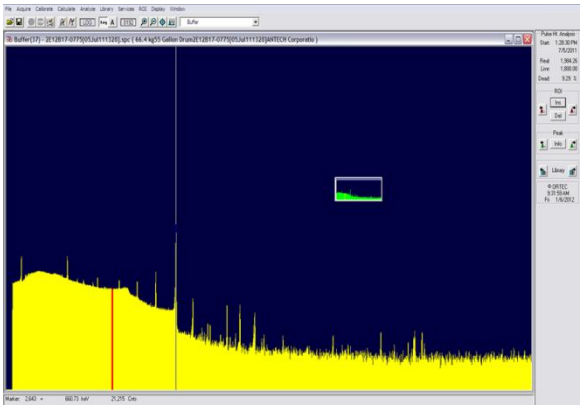
knowledge documentation identifies the presence of this radionuclide in the original waste population. The acceptable knowledge documentation is also very useful in identifying the relative location of waste drums containing this particular isotope and is effective in establishing a relative ratio for U-233/U-232 because the U-232 fraction is relatively small and the isotope activity based upon measured gamma ray signatures is not easily quantified. The population of drums with quantifiable amounts of U-233 can be subdivided into two additional populations. For approximately 50% of the measured U-233 containing drums, a significant presence of Th-232 is identified. Th-232, over a specified activity, is a reportable waste constituent for samples measured under contract No. 40245. Nearly half of the measured drums do not show significant Th-232 signatures.





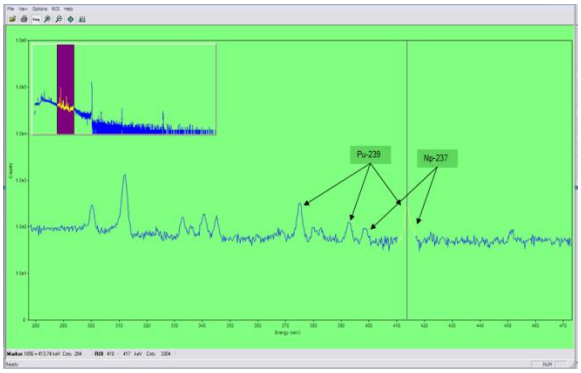
Figures 8a and 8b. U-233 indicator @ 440 keV, Th-232 indicators @ 969 and 911 keV

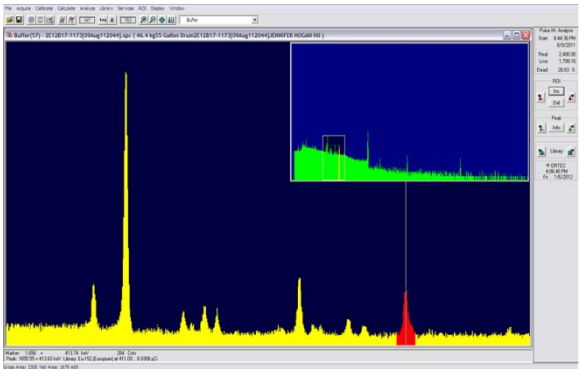




Figures 9a and 9b. High Cs-137, U-233 indicator @ 440 keV, Th-232 indicators @ 969 and 911 keV

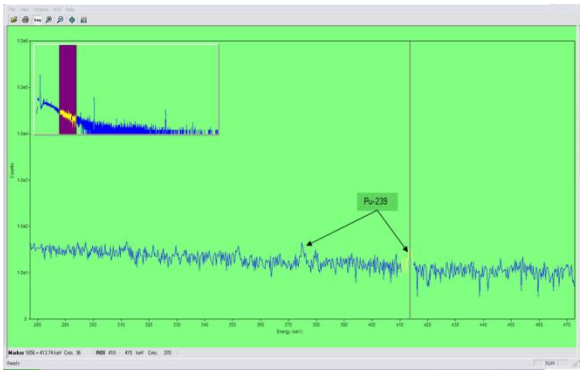
Samples that have a significant TRU radionuclide signature and are characterized as TRU waste drums comprise approximately 30% of the measurements for this contract. These samples usually report a significant isotopic signature for Pu-239 and Am-241, but other isotopes, both TRU and non-TRU, are identified and quantified in these samples. The presence of U-235 and U-238 isotopes is observed in many of the drums in this population and the TRU radionuclide, Np-237, is also routinely observed. The automated analysis of these samples is quite effective; however, the analyst must be cognizant of interferences induced by the presence of these isotopes. It is incumbent upon the analyst to confirm that appropriate corrections are performed in the analysis software.

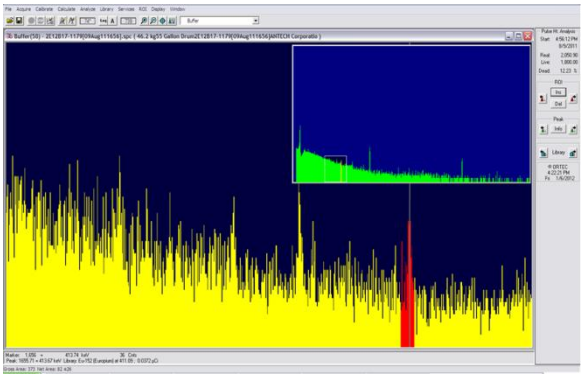




Figures 10a and 10b. Pu, Am, Np, U present; TRU characterization

Samples that do not have a substantial TRU radionuclide signature and are characterized as LLW comprise approximately 65% of the measurements for this contract. These samples are generally quite benign, isotopically, and rarely produce spectra with significant gamma ray signatures. Non-TRU isotopes, such as Co-60, Cs-137, Eu-152, Eu-154, Tl-208, Bi-214, Pb-214, and Th-232 are routinely identified, but do not approach the waste reporting limits for this contract. Occasionally, Pu-239 and Am-241 activities are identified and reported, but the summed activity for these isotopes, relative to the mass of the drum is less than 100 nCi/g. The isotopes of uranium are also prevalent in this population and the activities for these isotopes are reported; however, since these are not TRU isotopes the samples are usually characterized as LLW.





Figures 11a and 11b. Pu, Am, and U present; LLW characterization

Drum Statistics

Recovered drums were bagged and placed in the north drum staging location. All drums, exclusive of high dose drums due to the stated ALARA considerations, are weighed and measured on the ANTECH gamma ray assay equipment. This measurement protocol allows for an accurate assessment of the sample's gross weight and ensures that a gamma ray isotopic spectrum of the sample is collected.

NDA measurements for Contract 40245 commenced in February 2011 and were completed in August 2011. For this period of 189 days, from the first measurement batch to the final measurement batch, 64 measurement batches were compiled. Each batch is associated with a measurement day so that 64 batches correspond to 64 measurement days during the 189 days

(weekends and holidays are not excluded) of the measurement program. Of the 1514 waste drums measured 1426 of these drums, 94.2%, were resolved strictly by gamma ray analysis.

Drum measurement statistics relative to days of the week are tabulated in Table 1. The frequency of measurement days and the overall productivity for these measurement days are noted in the following table:

Day	Frequency	Samples	Samples/Day
Monday	11	200	18.2
Tuesday	16	451	28.2
Wednesday	12	271	22.6
Thursday	13	364	28.0
Friday	9	169	18.8
Saturday	1	24	24.0
Sunday	2	35	17.5
Total	64	1514	23.7

Table 1. Drum measurement statistics for days of the week during Contract No. 40245 for operation of the Gamma Mobile Assay Laboratory (G-MAL)

The reported “Frequency” identifies the number of days during the measurement period when measurements were performed; it does not identify the number of shifts performed for that measurement day. Note that two and three shift operations were not uncommon on Tuesdays and Thursdays and this can be seen in the statistics for those days. The “Samples/Day” is simply the quotient of the number of “Samples” divided by the “Frequency” or number of days and is the average number of drums for that day of the week. During the measurement campaign the G-MAL was never the limiting factor for throughput as it has 4 available HPGe based far field measurement stations. Factors that commonly limited drum measurement throughput were weather, manpower resource limitations (drum recovery and measurement shared staff) and limited availability of drums due to the difficulties of drum recovery discussed earlier.

CONCLUSIONS

This paper describes the process of radioactive waste drum recovery and drum assay for the purpose of LLW-TRU classification and segregation and of LLW drum characterization for final disposition conducted by CHPRC and ANTECH in the 200 Area West and 200 Area East at the Hanford Site. Issues relating to drum recovery in the 200 Area East burial grounds have been

described. The assay equipment has been described as has the methods and characteristics of spectroscopic data analysis and review, which is a critical part of the overall assay process.

For the remaining drums that are classified as LLW, a full assay and waste characterization is provided by the ANTECH assay systems. The initial classification and assay result for all drums is provided immediately and is followed by electronic data packages within 48 hours. These are provided by ANTECH SMEs based in Westminster to CHPRC waste disposition staff on site at Hanford. The data packages are subject to SME analysis and independent technical review and contain complete characterization data.

High drum throughput and reliability have been provided by the ANTECH automated far-field HPGe based Gamma Mobile Assay Laboratory (G-MAL) which has achieved drum measurement rates in excess of 60 drums in an 8 hour shift. The G-MAL is transportable and can be redeployed from site to site, where it requires only electrical power. Detector re-calibration is performed annually and can be undertaken in the field using low activity calibrated sources provided by ANTECH. The equipment is simple to use and has been operated by locally trained CHPRC operations staff with an ANTECH supervising operator present only part of the time when multi-shift operation takes place.

At the start of the first contract in 2004 the G-MAL (without automatic conveyor) was designed, built and deployed to site in a remarkable 31 days following contract placement. Following a period of four months of manual drum loading, the automatic conveyor was installed. With the conveyor and a supply of input drums, the entire measurement and drum handling process is automated.

The ANTECH Neutron Mobile Assay Laboratory (N-MAL) was designed, assembled and deployed to site within 120 days following contract placement in 2009 including the acquisition, testing and installation of the neutron generator for active neutron measurements. It can measure both drums and boxes using both active or passive neutron analysis. It has been used successfully to classify indeterminate drums (high gamma ray activity) as LLW or TRU.

In excess of 7,500 waste drums have been successfully assayed and sentenced since 2004 by the ANTECH NDA systems at Hanford. Of the total number of drums measured, between 25% and 30% were found to be TRU and these drums have been segregated for a further TRU processing.

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1. Currie, L. A. (1968). "Limits for Qualitative Detection and Quantitative Determination, Application to Radiochemistry", Analytical Chemistry 40(3):586-593.