NDA Challenges during Cleanup of the Hanford 618-10 Burial Ground Trenches – 15542

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ABSTRACT

The 618-10 Burial Ground on the Hanford Reservation was an early trench waste site used for the disposal of waste from the 300 Area at Hanford. Much of this waste consisted of residues from evaluations of failed fuel as well as various process and separations studies associated with the Department of Energy (DOE) mission which were performed in the 300 Area between March 1954 and September 1963. Much of this waste contained significant concentrations of fission products as well as Transuranic (TRU) radionuclides. Waste forms consist of drums and boxes which were disposed of in the trenches. Many of the drums are shielded containers, cemented sludge, or cemented debris. There are no identifying marks on the containers to correlate them to a specific waste stream. In addition there is some waste in the trenches which came from other locations outside of the 300 area.

Cleanup of the 618-10 burial ground is covered under the Washington Closure Hanford (WCH) contract. Due to the proximity of the 618-10 site to the Columbia River, waste is being removed from the 618-10 site, stabilized, and disposed of into either the Environmental Restoration Disposal Facility (ERDF) if the containers are determined to be LLW or transferred to the Hanford Central Waste Complex (CWC) if the containers are suspected to be TRU waste. In order to determine the correct disposal path Non Destructive Assay (NDA) must be performed on all of the containers excavated from the trenches.

The NDA of these containers is performed using germanium gamma spectroscopy with an In Situ Object Counting System (ISOCS) and with a neutron slab counter. Due to the challenges associated with these difficult waste streams, matrices, and lack of information to associate these containers with a particular process, the NDA program has had to develop a more robust process to ensure the correct disposition of the waste.

INTRODUCTION

The 618-10 Burial Ground on the Hanford Reservation was an early trench waste site used for the disposal of waste primarily from the 300 Area at Hanford. It accepted waste from March 1954 until September 1963. The 618-10 burial ground contains both trenches, which were used for LLW and some shielded high dose waste, and Vertical Pipe Units (VPU) which were used for disposal of non-shielded high dose rate waste. Much of the waste was from evaluations of failed fuel as well as residues from research on various extraction and separations processes. Remediation of the 618-10 burial grounds is a part of the Washington Closure Hanford (WCH) contract. This paper only relates to the NDA efforts associated with characterization of waste from the trenches. The trench waste included boxes, drums, and other items, including glove boxes.

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HISTORY

Drums

Historical information indicated that drums were primarily 30 gallon or 55 gallon drums. Some drums were strictly debris, although for drums containing higher dose rate materials drums may have had up to a 6 inch concrete liner and in some cases a $1^{\circ} - 3^{\circ}$ internal lead liner. There were also many drums containing sludges from solidified radioactive liquids, and debris mixed into concrete.



Examples of Concrete drums and debris drums

Inventory

Prior to the start of remediation, process history and available records were used to develop an expected inventory for the waste drums which were removed from the trenches. The primary source of waste which was expected was residues and fines from failed fuel evaluations of elements removed from the single pass reactors. This waste was typically disposed of in shielded drums due to the high fission product concentrations. A second source of waste was from experiments to develop the various extraction and separations processes. These included:

- PUREX the plutonium uranium extraction process
- REDOX the reduction oxidation process
- Th-232 separation from campaigns to produce U-233 material
- Np-237 separation from campaigns to produce Pu-238 for radioisotope thermoelectric generator (RTG) applications
- Sr-90 separated from the PUREX feed also for RTG applications

Other waste included significant quantities of uranium oxide and uranium metal from the production of fuels for the single pass reactors.

Based on the historical information a set of scaling factors were developed based on ORIGEN runs of fuel from the N reactor.

INITIAL CHARACTERIZATION WORK

Following excavation, the retrieved drum would be placed into an overpack drum, and then sent for characterization. During the initial characterization work a screening was performed using an ORTEC Detective to perform a qualitative identification of the primary nuclides present in the drum. At the same time a qualitative neutron slab measurement was performed to check for significant quantities of TRU material. Drums where only U and/or Th was detected did not require further characterization. All other drums were assayed for 15 minutes using an ISOCS system to obtain a quantitative measurement. Measured nuclides from the ISOCS measurement were applied to the scaling factors to develop the radionuclide inventory for the drum. For high density or shielded drums, Cs-137 was frequently the only radionuclide detected.

In order to ensure conservatism a procedure was developed which required the most conservative model to be chosen based on the weight of the drum. For heavy drums, this was typically the concrete shielded or lead lined concrete shielded drum model. The only criteria which could be used to select a less conservative model was based on the results of a Line Activity Consistency Evaluation (LACE) analysis. LACE is a tool which is available in the ISOCS software (See Figure 1). Typically with the 15 minute assay time, there were not adequate peaks available to perform a LACE analysis.

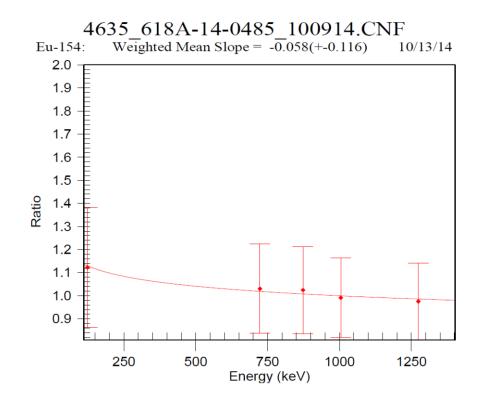


Figure 1: Typical LACE analysis Curve

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In several cases when the 59.5 keV peak from Am-241 was detected, the conservative model produced an unrealistically high activity for the Am-241. Because of this an assumption was made that the Am-241 was caused by surface contamination on the inner drum. The assumption of surface contamination was later called into question and the characterization program was halted until the process could be fully validated.

MAKING THE NDA PROGRAM MORE ROBUST

Validation of the ISOCS System

As a part of the program evaluation an NDA Requirements Document which was in use under the Plateau Remediation Contract (PRC) at Hanford was applied to the WCH NDA program. A key part of this document was the requirement to validate the measurement capability of the NDA instrument to detect and quantify TRU nuclides to a reasonable precision and accuracy.

A test plan was developed to test the ability of the ISOCS system to measure TRU nuclides at or near the 100 nCi/g TRU/LLW sorting criteria when placed into a sludge type of matrix or into a shielded concrete drum. The test was setup at the Hanford Waste Receiving and Processing (WRAP) facility since there were certified plutonium standards available.

Based on an initial review of the existing ISOCS data, it was determined that the system could not detect the Pu in a shielded drum using a 15 minute count time. Therefore the assay time was increased to 2 hours prior to performing the test. Results from the test using the 2 hour count time documented that the ISOCS system could be used for TRU/LLW sorting with a 2 hour count time under most circumstances.

With this qualification in place the NDA program was restarted. Although the 2 hour count time greatly increased the ability to perform a LACE analysis on many drums and revise models down to uniform or slightly shielded sludge models, there were still many drums where the Compton scatter from fission product nuclides inhibited the ability of the ISOCS system to detect key Pu lines. Therefore many drums were classified as suspect TRU based on the MDAs for Am-241 and Pu-239.

Validation of the Hanford Slab Counter

To reduce the number of suspect TRU drums it was decided to perform a similar qualification on the Hanford neutron slab counter (HSC) at the WRAP facility. The HSC operates in a singles counting mode. Therefore spontaneous fissions cannot be discriminated from alpha-n reactions which may occur in the waste, spallation neutrons, or the spontaneous fission of U-238. Therefore the validation test was modified from requiring a specific accuracy, to providing a bounding value for the measurement.

Once the system was set up at the WRAP facility, it was determined that the neutron background count rate was 6-7 times higher than the background rate at the 618-10 site. Therefore Pu masses for the validations testing were increased to meet the higher detection level at the WRAP facility. With the high gram levels in place, the HSC passed both the precision and accuracy test for both the sludge and the concrete shielded drums.

With both systems qualified for quantitative measurements, a ranking system was put into place for applying the waste stream scaling factors to the assay results. The following is a table of the ranking:

- 1. ISOCS Measured Pu-239 activity
- 2. ISOCS Measured Am-241 if no Pu-239 is present
- 3. HSC Measured provisional Pu if Am-241 and Pu-239 are both MDAs
- 4. ISOCS measured Cs-137 if it produces a higher TRU value than the HSC results.

Having the combination of these two tools has greatly reduced the number of drums being classified as suspect TRU due to a lack of adequate measurement data.

Interesting correlations

On the HSC counter there are several phenomena that can cause elevated values. In several cases these are somewhat quantifiable and can be used as a part of the evaluation.

- 1. For uranium oxide drums a HSC value of 1 cps correlates to 4.7 kg of U-238. This has become useful as a warning tool since a few of the drums with significant kg quantities of UOx have been known to spontaneously combust when emptied for stabilization. A high HSC value can be used as a warning indicator.
- 2. Drums which contain an internal lead liner have an elevated HSC result due to spallation. It is known from historical data that there are drums with 1", 2" and 3" lead liners. Identification of these is somewhat predicable based on the drum weight; however the HSC result increases by approximately 0.4 cps for each inch of thickness in the lead liner.
- 3. There is also a correlation between the alpha-n present and the quantity of Th-232 present in the drum, however there is significantly high variation on this data. In general 100uCi of Th-232 produces a count rate of approximately 1 cps.

CURRENT OPERATIONS

With the current process in place the debris and sludge drums are typically dispositioned based on the ISOCS assay results. For drums with adequate quantities of Pu, there is usually good concurrence between the ISOCS and the HSC results.

For the shielded drums, containing residues from the failed fuel examination, the Compton scatter from the Cs-137 typically masks the ability to detect any TRU nuclides using the ISOCS system. The HSC data is normally used to disposition the drums and typically has a low enough detection level to allow these drums to be dispositioned as LLW. Even in cases where there is a lead lining the increased drum weight offsets the higher neutron result caused by the spallation neutrons so that the final result still ends up as LLW.

There are still a few drums which are pushed into a suspect TRU category due to high MDA results for both the ISOCS and HSC counters, but most of the drums can now be properly dispositioned.

Some Interesting Data

It was known that there were a number of drums containing Sr-90 which were buried in the 618-10 trenches. These drums have been easy to identify due to their characteristic Bremsstrahlung continuum as show in figure 2 below. The continuum also tends to mask the ability to detect the lower energy TRU nuclides with the ISOCS system. Although these drums can be identified, they cannot be quantified. However since the Sr-90 concentration limit at the ERDF facility is very high, these drums can be accepted for disposal. Work to develop a pseudo-quantitative technique is being studied.

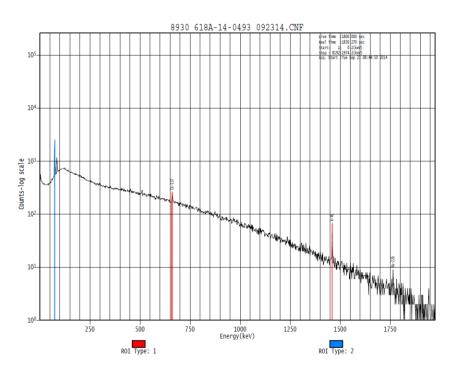


Figure 2: Sr-90 Bremsstrahlung Spectrum

DEVELOPMENTS STILL IN PROCESS

In some cases the 59.5 keV peak for the Am-241 is detected in a high density drum; however there is not adequate data to perform a LACE analysis. For most of these drums the MDA for the 722 keV line from Am-241 is several orders of magnitude lower than the measured value from the 59.5 keV peak. This is an obvious indicator that the ISOCS model is too conservative and should be reduced.

The site is in the process of upgrading the ISOCS data acquisition software to Canberra NDA2000. The software upgrade will provide several benefits for both the data collection and analysis once it is completed. Several types of data collection errors will be eliminated with the new software, and for some types of drums the data analysis and processing can be automated.

PROBLEMS NOT ADDRESSED YET

There are a few drums which have been set aside due to high dose rates. Most of these drums may be able to be counted on the ISOCS system using a combination of shielding and distance. However the overall dose rate in the characterization area may create an ALARA issue also. There is also one drum which may have a high enough dose rate to affect the HSC counter. This will need to be evaluated during the measurement process. Addressing these issues will help to prepare the characterization program for a larger number of high dose rate drums which will come out of the VPU remediation program.